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Isotopes & Radiation Technology

ISRTA$_I$ 9(2) 129–252 Winter 1971–1972

I Isotope Production and Development

129 **Isotope Production in Commercial Power Reactors** *P. M. Wood*

133 **The Chemistry of the Electromagnetically Separated Stable Isotopes** *R. E. Greene*

150 **Xenon-128 Production Process Development (D. W. Haase)** *Summarized by R. E. Greene*

150 Experimental

154 **Evaluation of Nuclear Reactions that Produce ^{123}I in the Cyclotron**

154 ^{123}I Production Reactions

154 Reactions Investigated

159 Conclusions

160 **Instant ^{99m}Tc Compounds** *W. C. Eckelman and P. Richards*

160 Preparation of ^{99m}Tc–DTPA

160 Preparation of ^{99m}Tc–HSA

II Isotope Technology Development

162 **Radioanalytical Methods Applied to Air–Sea Interaction Studies** *P. E. Wilkniss, D. J. Bressan, and R. E. Larson*

162 Activation Analysis

163 Transport of Particulates from the Ocean into the Atmosphere—A Laboratory Investigation with Radiotracers

166 Conclusions

167 **Automatic Industrial Activation Analysis System for Slurries Using a Radioisotope Neutron Source** *Kenneth R. Blake, John B. Ashe, Peter F. Berry, and John B. Nelson*

167 Technique

168 System Description

171 Input Requirements

171 Economic Justification

171 System Performance

172 Conclusions

173 **Energy Dispersion X-Ray (EDX) Analysis in the Nonferrous Mining Industry** *A. P. Langheinrich, J. W. Forster, and T. A. Linn, Jr.*

173 Equipment

176 Applications

180 Summary

182 **Radioisotopic Monitor for Rime Accumulation** *W. B. Fowler*

183 **In Situ Elemental Analysis for Oceanographic Applications** *J. H. Reed and J. W. Mandler*

III Food Irradiation

186 **Status of Approval of Irradiated Food Products for Human Consumption**

188 **Wholesomeness of Irradiated Food, Especially Potatoes, Wheat, and Onions** *Summarized by F. E. McKinney*

188 Data Extrapolation
188 Mutagenicity and Cytotoxicity
188 Investigations Required for Assessment of Wholesomeness
189 Evaluation of Irradiated Foods
189 Irradiated Wheat
191 Irradiated Potatoes
192 Irradiated Onions
193 Future Research
193 Recommendations

194 **Effects of Low-Level Irradiation on the Preservation of Fruit—A Seven-Year Summary** *R. A. Dennison and E. M. Ahmed*

194 Citrus
196 Peaches
198 Mangoes
198 Tomatoes
199 Microbiological Studies
199 Bibliography

IV Process Radiation Development

202 **New Aspects in the Field of Gamma-Irradiation Plants** *H. Christen*

202 "Double-Cell" Laboratory Irradiation Plant
203 "Unicell" Industrial Plants

204 **Commercial Gamma-Irradiation Plant in Sweden** *C. G. Crawford*

204 Plant Description
206 Plant Operation

208 **Ezeiza Semi-Industrial Irradiation Plant**

V Source Development

209 **Recent Developments in Radioisotope Neutron Sources** *R. W. Tolmie*

209 (α,n) Sources

VI Isotopic Heat and Power Development

215 **Comparison of Radioisotope-Fueled Energy Sources for a Prosthetic Device** *Joel D. Hixson*

216 Thermoelectric Conversion
218 Thermionic Conversion
219 Betavoltaic Conversion
220 Comparison of Fuels
221 Overall Comparison

222 **Safety Features of French Cardiac Pacemaker Fueled with ^{238}Pu** *R. Berger, R. Boucher, B. Jampsin, and M. Hot*

223 Construction Requirements
223 Safety Requirements

230 **^{60}Co Use in Thermoelectric Generators** *Summarized by Martha Gerrard*

230 U. S. Studies
231 MAPLE Generators
232 GICODYNE Generator

235 **Comparative Evaluation of Power-System Types for Underseas Applications** *Robert E. Wimmer*
235 Power-System Parameters
237 Mission Parameters
237 Conclusions
238 **SNAP-21**
238 System Description
238 Safety System
238 Operational Performance

VII Applications in Life Science

240 **Automated Radiometric Detection of Microorganisms** *Frank H. DeLand, M.D.*
240 Method
240 Results
244 **Radioisotopic Technique for Studying Biological Slime-Treatment Processes** *George W. Reid*
244 Instrument

VIII Miscellaneous

247 **Availability of Isotopes and Services**
248 **AEC Activities**
248 **General**

COVER: Installation of radioisotopic rime-accumulation monitor. USDA Forest Hydrology Laboratory photograph. See page 182.

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Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

Insofar as feasible all references are checked by the editors, using the library facilities of Oak Ridge National Laboratory, Oak Ridge Associated Universities, and the University of Tennessee. Occasionally, however, the cited material is not available; such cases are identified by an asterisk (*).

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Heat and Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

P. S. Baker, R. H. Lafferty, Jr., Martha Gerrard, R. E. Greene, F. E. McKinney, and Helen P. Raaen, *Editors*
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Section I

Isotopes and Radiation Technology

Isotope Production and Development

Isotope Production in Commercial Power Reactors

By P. M. Wood*

Supplementary Keywords: *reactor; production, radioisotope; ^{60}Co; ^{238}Pu; ^{237}Np, ^{241}Am; ^{243}Am; ^{242}Cm; ^{244}Cm.*

Abstract: *Isotope production in power reactors is now limited to ^{60}Co, but ^{238}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, and ^{244}Cm are potential products. Calculated production quantities are presented.*

The major production of isotopes in domestic power reactors is currently limited to ^{60}Co production by Neutron Products, Inc., and Consumers Power Company. Neutron Products, Inc., has delivered over 1 million Ci produced in the Big Rock Point Reactor and has sold some 4 million Ci for delivery over the next 3 years. General Electric Company produces ^{60}Co in the General Electric Test Reactor and may in the future produce it in commercial reactors. The first Canadian Pickering power reactor will start up in 1971, and by 1975 the ^{60}Co production from the four Pickering reactors may be 12 to 15 MCi/year.

The main difficulty with ^{60}Co produced in a light-water reactor (LWR) is its low specific activity; it is difficult to obtain specific activities of more than 100 Ci/g. Although low specific activities are useful in many processes, such as sterilization of medical supplies and irradiation of wood, some applications, such as medical therapy, require high specific activities ($\geq$400 Ci/g). At present, high-specific-activity material must be obtained from sources other than power reactors. Commercial suppliers are Atomic Energy of Canada Ltd. and the General Electric Company.

*Union Carbide Corporation, AEC Combined Operations Planning (AECOP), P. O. Box S, Oak Ridge, Tenn. 37830. This is a somewhat abbreviated version of a paper presented at the American Nuclear Society Meeting, Los Angeles, Calif., July 1, 1970.

Another isotope of interest that can be produced in a power reactor is ^{238}Pu, which can be produced by irradiation of ^{237}Np or ^{241}Am. Both target materials can be recovered from discharged fuel. Unfortunately, the ^{238}Pu produced by ^{237}Np irradiation in an LWR spectrum is contaminated by ^{236}Pu, which is present at concentrations of the order of 10 ppM. The ^{236}Pu decays with a 2.85-year half-life to ^{232}U with its well-known chain of radioactive daughters. Plutonium-238 containing ^{236}Pu may have some use as a radioactive heat source in applications that do not require low gamma fields; it is possible that ^{238}Pu contaminated with ^{236}Pu could be used in such an application for 10 years (four half-lives) and then be reprocessed, to remove ^{232}U and its daughters, for use as medical-grade ^{238}Pu ($<$0.3 ppM ^{236}Pu). This is probably not a very sound investment scheme since the cost of converting ^{237}Np to medical-grade ^{238}Pu in a Canadian heavy-water reactor is not likely to be more than twice the cost of making contaminated ^{238}Pu in an LWR. In addition to this poor return on investment (about 5%), the market for medical-grade ^{238}Pu is, at best, uncertain 10 or more years from now.

The production of ^{238}Pu by irradiation of ^{237}Np was studied extensively 10 years ago by Neutron Products, Inc., and Dow Chemical Co. The project was given up for lack of a firm market for ^{238}Pu. Neutron Products, Inc., holds several patents in the field.

Recently the AEC sponsored a program for Southern Nuclear Engineering Company (SNE) to carry out a study of ^{238}Pu production by ^{237}Np irradiation in LWRs. Calculations by both SNE and AEC Combined Operations Planning (AECOP) show that sizeable amounts of ^{238}Pu can be produced in this manner. For example, neptunium targets could be placed in the water holes of a pressurized-water reactor (PWR) in a

mechanical configuration similar to that used for burnable poison rods.

Again, the major problem with ^{238}Pu produced by ^{237}Np irradiation in PWRs is the ^{236}Pu contamination. To resolve this problem, a 2-year experimental program* has been planned, calling for irradiation of 10 ^{237}Np samples diluted with zirconium oxide to five different concentrations to study the effect of resonance self-shielding. A power reactor for the irradiations has not been chosen, but several operators have expressed interest in participating in the program. It would be desirable to use a core with zirconium-clad fuel rather than a core with stainless-steel cladding, as most reactors in the future will use zirconium cladding. Unfortunately, most of the operating PWRs have stainless-steel-clad fuel. If a core with stainless-steel cladding is used for the experiments, the field of iron capture gammas from the stainless steel, giving rise to (γ,n) reactions in ^{237}Np, will make the program less meaningful. The postirradiation program would call for a spectrographic determination of plutonium isotopic concentrations as well as an analytical evaluation.

The following AECOP results illustrate the potential of ^{238}Pu production in the commercial power-reactor industry. Figure 1 shows the concentration of ^{237}Np and ^{238}Pu in a dilute target vs. irradiation time in a PWR of the Diablo Canyon type. Figure 2 shows the percentage of ^{238}Pu in the plutonium mixture as a function of time. A 1-year irradiation would convert 24% of the ^{237}Np to ^{238}Pu with less than 15% dilution by the other plutonium isotopes.

If the by-product ^{237}Np produced in commercial reactors could be recovered with a yield of 90% during fuel processing, the quantities of ^{237}Np shown in Table 1 would be available. If this ^{237}Np should be irradiated in LWRs, as described above, the quantities of ^{238}Pu (with approximately 10 ppM ^{236}Pu contamination) produced would be as shown in Table 2.

An alternative way of producing ^{238}Pu in commercial power reactors is by the ^{241}Am chain. Upon irradiation, the ^{241}Am, which is produced by ^{241}Pu decay, absorbs a neutron to produce ^{242}Am, which beta decays to ^{242}Cm. Curium-242 alpha decays with a 163-day half-life to ^{238}Pu.

The AECOP projections of the availability of ^{241}Am, ^{243}Am, ^{242}Cm and ^{244}Cm, and medium-grade ^{238}Pu are shown in Table 3. The $^{242}Cm/^{244}Cm$

*Note added in proof: Eight ^{237}Np targets of four different concentrations were loaded in the Haddam Neck Reactor (Connecticut Yankee) in May 1971 and will be irradiated until the next refueling in the fall of 1972.

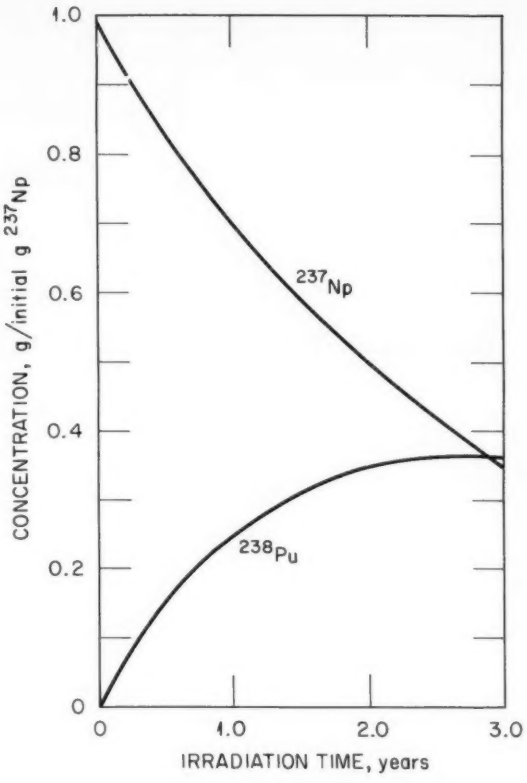

Fig. 1 Concentrations of ^{237}Np and ^{238}Pu in target as a function of irradiation time in a PWR (80% load factor).

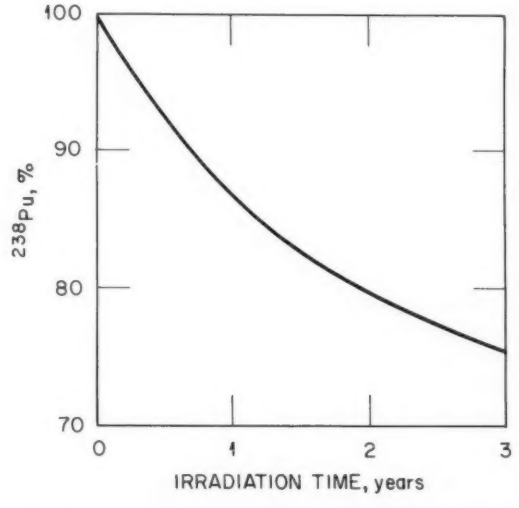

Fig. 2 Concentration of ^{238}Pu in plutonium mixture produced by irradiation of ^{237}Np target in a PWR as a function of time (80% load factor).

Table 1 ^{237}Np Production in Domestic Power Reactors Through 1980

Calendar year	^{237}Np,* kg
No Plutonium Recycle	
1970	10
1971	21
1972	34
1973	84
1974	179
1975	394
1976	556
1977	674
1978	777
1979	956
1980	1135
With Plutonium Recycle Starting in 1976	
1979	873
1980	1025

*10% loss during fuel processing assumed.

Table 2 ^{238}Pu That Could Be Produced by Irradiating Available ^{237}Np in Commercial Power Reactors

Calendar year	^{238}Pu, kg	Fiscal year	^{238}Pu, kg
No Plutonium Recycle			
1974	10	1974	
1975	32	1975	20
1976	83	1976	50
1977	149	1977	117
1978	229	1978	191
1979	308	1979	266
1980	394	1980	350
1981	493	1981	445
		1982	548
With Plutonium Recycle Beginning in 1976			
1980	380		
1981	463	1981	425
		1982	504

Table 3 Amounts of Isotopes Available, per Fiscal Year, from Domestic Power Reactors

Isotope	Case 1. No Plutonium Recycle								
	1972	1973	1974	1975	1976	1977	1978	1979	1980
^{241}Am	3.36	5.34	14.8	26.2	63.2	86.5	102.3	119.3	142.1
^{243}Am	2.82	4.27	10.0	19.8	52.1	69.8	93.8	111.8	127.2
^{242}Cm									
At reactor discharge	0.52	0.82	2.00	3.70	9.12	12.0	17.8	21.7	25.0
After recovery	0.24	0.38	0.92	1.70	4.20	5.52	8.19	9.986	11.51
^{244}Cm	0.71	2.14	2.26	4.70	13.9	18.3	26.6	33.7	36.8
^{238}Pu (medical grade)									
1-year decay		0.19	0.29	0.72	1.33	3.28	4.31	6.34	7.79
2-year decay			0.23	0.35	0.87	1.61	3.97	5.22	7.75

Isotope	Case 2. Plutonium Recycle 1 Year After Available		Case 3. Plutonium Recycle 2 Years After Available	
	1979	1980	1979	1980
^{241}Am	177.3	253.3	134.6	188.5
^{243}Am	195.5	287.7	133.9	194.1
^{242}Cm				
At reactor discharge	41.2	62.4	26.9	40.6
After recovery	18.96	28.72	12.38	18.68
^{244}Cm	114.2	191.4	55.0	101.2
^{238}Pu (medical grade)				
1-year decay	6.34	14.80	6.34	9.66
2-year decay		7.75	5.22	7.75

mixture could be recovered during spent-fuel processing and allowed to decay.

Alternatively, the $^{241}Am/^{243}Am$ mixture could be irradiated as a target in a manner similar to that described above for ^{237}Np. The $^{242}Cm/^{244}Cm$ mixture produced would in turn be allowed to decay to produce ^{238}Pu. The amount of medical-grade ^{238}Pu that could be made in this way is shown in Table 4. The ^{238}Pu produced would be diluted with 15 to 20% ^{242}Pu produced by K capture in ^{242}Am. The spontaneous fissions in ^{242}Pu generate radiation equivalent to about 0.1 ppM ^{236}Pu.

After a suitable time for decay of ^{242}Cm to ^{238}Pu, the ^{238}Pu and curium isotopes would be separated; the ^{244}Cm and higher curium isotopes would be available for other applications, such as

Table 4 Amounts of ^{242}Cm and ^{238}Pu Available from Irradiation of ^{241}Am in LWRs

1-Year-Target Irradiation; ^{241}Am Recycle After 6 Months

Isotope	Amount, kg										
	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983
^{242}Cm											
At target discharge	0.73	1.34	3.69	6.98	16.03	24.05	31.08	39.07	48.04		
After 1-year decay		0.16	0.28	0.78	1.48	3.44	5.18	6.74	8.41	10.02	
After 2-year decay			0.03	0.06	0.16	0.31	0.73	1.10	1.43	1.78	2.17
^{238}Pu											
At target discharge	0.74	1.35	3.71	6.99	16.04	24.06	32.00	40.00	48.06		
After 1-year decay		1.32	2.41	6.62	12.05	29.02	43.09	57.01	71.03	86.08	
After 2-year decay			1.44	2.63	7.23	13.07	31.09	47.09	62.04	77.09	98.04

Table 5 Transplutonium Isotope Production in a PWR

Isotope	1-year exposure	2-year exposure	3-year exposure	4-year exposure	5-year exposure
	^{243}Am, g/initial kg				
^{243}Am	619	383	237	147	91
^{244}Cm	337	475	504	478	427
^{245}Cm	19.7	37.9	45.0	45.0	41.4
^{246}Cm	2.49	11.4	23.8	36.7	48.5
^{247}Cm	0.0227	0.208	0.635	1.26	2.00
^{248}Cm	9.04×10^{-4}	0.0179	0.0877	0.247	0.516
^{249}Bk	8.42×10^{-6}	2.74×10^{-4}	1.70×10^{-3}	5.45×10^{-3}	0.0124
^{249}Cf	2.23×10^{-6}	1.24×10^{-4}	9.91×10^{-4}	3.71×10^{-3}	9.34×10^{-3}
^{250}Cf	1.46×10^{-6}	8.34×10^{-5}	6.79×10^{-4}	2.57×10^{-3}	6.53×10^{-3}
^{251}Cf	2.94×10^{-7}	2.45×10^{-5}	2.33×10^{-4}	9.60×10^{-4}	2.56×10^{-3}
^{252}Cf	5.80×10^{-8}	1.13×10^{-5}	1.55×10^{-4}	8.84×10^{-4}	3.03×10^{-3}
	^{244}Cm, g/initial kg				
^{244}Cm	789	623	492	388	306
^{245}Cm	692	63.0	50.7	40.1	31.7
^{246}Cm	14.9	34.8	50.7	62.6	71.2
^{247}Cm	0.183	0.842	1.75	27.0	3.58
^{248}Cm	9.34×10^{-3}	0.0938	0.314	0.686	1.19
^{249}Bk	1.01×10^{-4}	1.62×10^{-3}	6.68×10^{-3}	0.0164	0.0306
^{249}Cf	3.06×10^{-5}	8.20×10^{-4}	4.30×10^{-3}	0.0120	0.0245
^{250}Cf	2.02×10^{-5}	5.56×10^{-4}	2.97×10^{-3}	8.41×10^{-3}	0.0172
^{251}Cf	4.45×10^{-6}	1.74×10^{-4}	1.07×10^{-3}	3.27×10^{-3}	6.99×10^{-3}
^{252}Cf	1.01×10^{-6}	8.56×10^{-5}	8.35×10^{-4}	3.53×10^{-3}	9.64×10^{-3}

^{252}Cf production. It is unlikely that ^{252}Cf would be produced in a commercial power reactor because of the relatively low thermal neutron flux characteristic of such reactors. A flux of the order of 10^{15} n/(cm^2)(sec) is required because of the ^{252}Cf production-chain "bottlenecks," which result from the low cross sections of ^{246}Cm and ^{248}Cm. It may, however, be desirable to produce curium isotopes up to ^{246}Cm by irradiation of ^{243}Am and ^{244}Cm in power reactors and then use the product curium isotopes as targets in a high-flux reactor such as the ORNL HFIR (High Flux Isotope Reactor) or the AEC production reactors at Savannah River. Table 5 shows the quantities of these curium isotopes that could be produced by irradiating 1 kg of either ^{243}Am or ^{244}Cm.

(MG)

The Chemistry of the Electromagnetically Separated Stable Isotopes

Supplementary Keywords: *enrichment, isotope; electromagnetic process and equipment; electromagnetic final chemistry; precipitation; ion exchange; volatilization; chemistry, inorganic; ^{106}Cd; ^{108}Cd; ^{110}Cd; ^{111}Cd; ^{112}Cd; ^{113}Cd; ^{114}Cd; ^{116}Cd; ^{35}Cl; ^{37}Cl; ^{113}In; ^{115}In; ^{102}Pd; ^{104}Pd; ^{105}Pd; ^{106}Pd; ^{108}Pd; ^{110}Pd; ^{39}K; ^{40}K; ^{41}K; ^{96}Ru; ^{98}Ru; ^{99}Ru; ^{100}Ru; ^{101}Ru; ^{102}Ru; ^{104}Ru; ^{180}Ta; ^{181}Ta; ^{46}Ti; ^{47}Ti; ^{48}Ti; ^{49}Ti; ^{50}Ti.*

Abstract: *In this fifth article of a series reviewing the chemical purification of stable isotopes enriched electromagnetically at Oak Ridge National Laboratory, the purification methods for isotopes of Cd, Cl, In, Pd, K, Ru, Ta, and Ti are given.*

This is the fifth article of a series[1-4] on the chemical purification of electromagnetically separated stable isotopes enriched at Oak Ridge National Laboratory. The general problems and methods associated with the handling and purification of isotopically enriched materials, and with their isotopic and chemical analyses, were described in the first article of the series, which also includes a description of the chemical processing methods for the enriched isotopes of Sr, Ba, Pb, Zr, and Hf. Processing methods for Cr, Pt, and Os were given in the second of the series, Ce in the third, and W in the fourth. Descriptions of the chemical purification methods for Cd, Cl, In, Pd, K, Ru, Ta, and Ti, which are presented in detail in USAEC Report ORNL-4583 (Ref. 5), follow. In the descriptions of the purification of isotopes of these various elements, a very brief summary precedes a more detailed presentation of the purification method. The values given for natural abundances of the isotopes are also from this reference.

Cadmium

Cadmium has eight naturally occurring isotopes with abundances as follows:

Isotope	Natural abundance, at.%	Isotope	Natural abundance, at.%
^{106}Cd	1.215	^{112}Cd	24.07
^{108}Cd	0.875	^{113}Cd	12.26
^{110}Cd	12.39	^{114}Cd	28.86
^{111}Cd	12.75	^{116}Cd	7.58

Both aluminum and copper water-cooled pockets are used for the collection of these isotopes in the electromagnetic separation process, and the purification procedures relative to the two types of pockets are different enough to warrant separate descriptions. In general, both procedures involve recovery of cadmium on an ion-exchange column, multiple precipitations as CdS, followed by a final precipitation as $CdCO_3$ and calcining to CdO, the stable inventory form. Plastic centrifuge tubes, beakers, covers, etc., are used when the cadmium is processed in alkaline solutions.

Purification of Cadmium Isotopes Collected in Aluminum Pockets. The detailed procedure for this purification is shown schematically in Fig. 1*a*. Removal of cadmium deposits from aluminum pockets is somewhat similar to that from copper pockets[1] and involves cleaning the outside of the aluminum pocket with 1 : 1

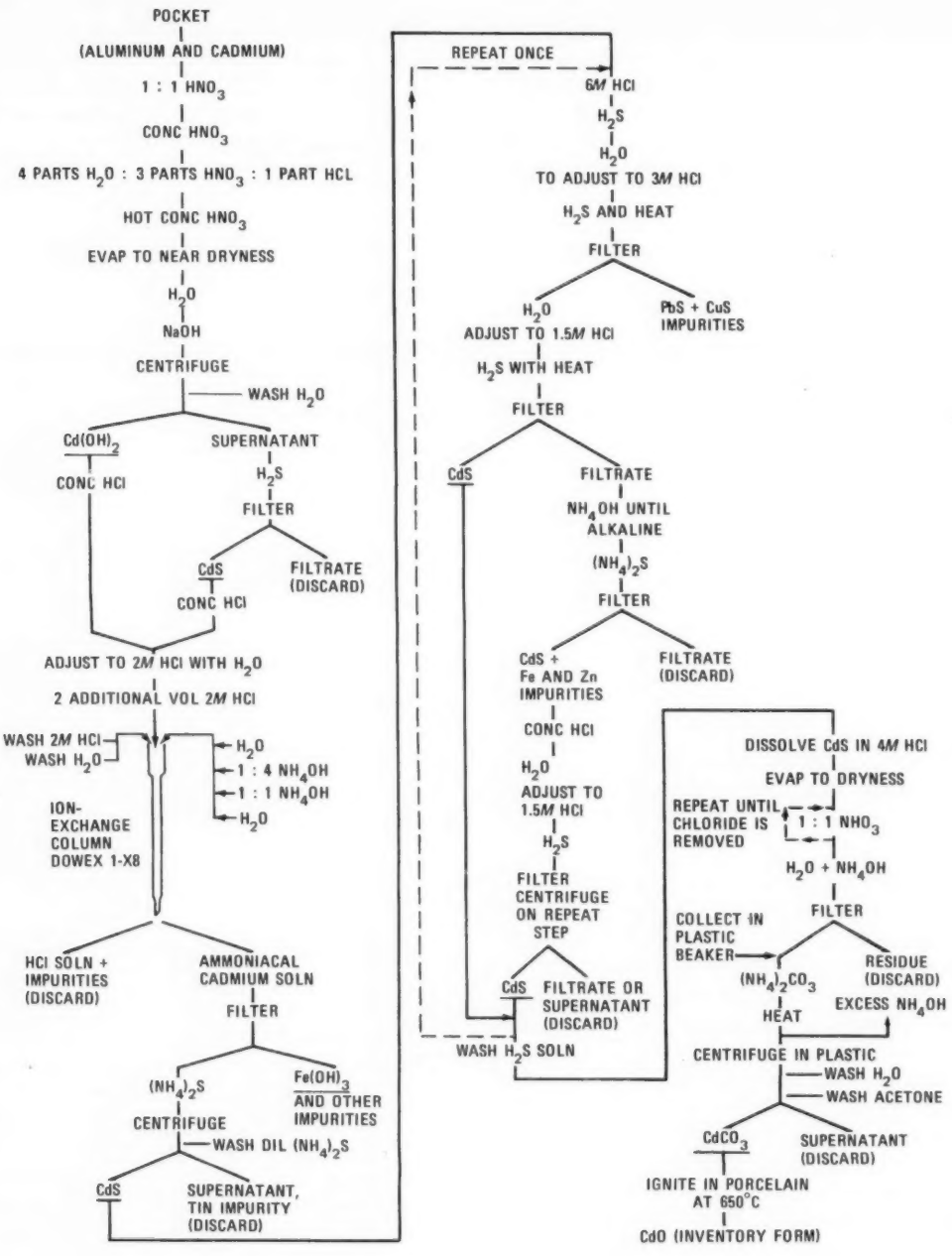

Fig. 1*a* Schematic diagram of the chemical purification procedure for cadmium isotopes collected in aluminum pockets.

HNO_3, removal of the protective tape covering the opening of the pocket, prying loose the adhering flakes of deposit, and transferring the loose material to a Pyrex dish. The deposit remaining within the pocket is leached with concentrated HNO_3 and then with a solution of 4 parts H_2O : 3 parts HNO_3 : 1 part HCl, followed by a final rinsing with hot concentrated HNO_3.

The loose solids and the leach solutions are transferred to a beaker and heated until the solids dissolve. The solution is evaporated to near dryness, and the residue is redissolved in water.

Cadmium is precipitated as $Cd(OH)_2$, with NaOH; most of the aluminum and zinc impurities are converted to the soluble aluminate and zincate, respectively. Cadmium hydroxide is separated by centrifuging and washing with water. The supernatant liquid is treated with H_2S to recover any cadmium remaining in solution. The recovered CdS is dissolved in concentrated HCl.

The $Cd(OH)_2$ is dissolved in a minimum amount of concentrated HCl and combined with the cadmium solution recovered from the above-mentioned sulfide precipitation. The resultant solution is adjusted to 2*M* HCl with water, and then to this volume of solution is added two additional volumes of 2*M* HCl.

In the next step cadmium is removed from solution by ion exchange, using a column of Dowex 1-X8 of Bio-Rad (or AR) quality, 100 to 200 mesh size, and in the chloride form. The size of the column is dependent on the amount of cadmium being processed—about 100 ml of wet resin for each 5 g of cadmium. The cadmium solution is poured through the column at a rate of ~20 ml/min; the column is then washed, first with 2*M* HCl and then with a small amount of water to remove excess HCl.

Cadmium is eluted from the column by washing with water, 1 : 4 NH_4OH, then 1 : 1 NH_4OH, and again with water (the column is reconditioned by washing with water and then 2*M* HCl). The ammoniacal solution is filtered to remove insoluble hydroxides, such as $Fe(OH)_3$, and then to this solution is added $(NH_4)_2S$. The cadmium sulfide precipitate is separated from the solution by centrifugation and washed with a dilute solution of $(NH_4)_2S$. Any tin impurity present will remain in the supernatant liquid.

The CdS is dissolved in 6*M* HCl and the solution saturated with H_2S, then adjusted to 3*M* HCl with water, and again saturated with H_2S. Sulfides of such impurities as lead and copper are coagulated by heating before their removal by filtration. (Cadmium sulfide will precipitate if its concentration is too high; 20 to 50 g of cadmium in 4 liters of solution is satisfactory.)

After separation of impurities as sulfides, the cadmium solution is again adjusted with water to give a solution 1.5*M* HCl, and H_2S is added while heating. The bulk of the cadmium is precipitated as CdS and recovered by filtration. The cadmium remaining in the filtrate is precipitated by addition of first NH_4OH until the solution is alkaline and then $(NH_4)_2S$. Any iron or zinc present is also precipitated. The solution is filtered and the cadmium recovered as the sulfide by dissolving in concentrated HCl and again precipitating with H_2S from 1.5*M* HCl solution.

The CdS precipitates are combined and dissolved in 6*M* HCl solution, and the complete sulfide precipitation procedure is repeated once again.

The CdS recovered from the second precipitation is centrifuged, washed with H_2S solution, and dissolved in 4*M* HCl. The chloride solution is evaporated to dryness, and then the residue is repeatedly evaporated with 1 : 1 HNO_3 until all chloride has been removed. The nitrate is dissolved in water containing enough NH_4OH to make the resulting solution alkaline and to complex the cadmium. Any precipitate present, e.g., iron and aluminum, is removed by filtration; the filtrate is collected in a plastic beaker (the concentration of cadmium should be ~50 g of cadmium/2 liters of solution).

Cadmium is now homogeneously precipitated as the carbonate by the addition of excess $(NH_4)_2CO_3$ solution and the slow removal of excess NH_4OH by evaporation. The precipitate is centrifuged and washed well, first with water and then with acetone. The solid is dried on a steam bath or with warm air to remove acetone and then transferred to a porcelain dish for subsequent ignition, starting at room temperature and increasing to 650°C. The resulting CdO is the stable inventory form.

Purification of Cadmium Isotopes Collected in Copper Pockets. The detailed procedure for this purification is shown schematically in Fig. 1*b*. The outside of the copper collector pocket is cleaned first with water and then with 1 : 1 HNO_3, followed by removal of the protective tape.[1] The isotope within the pocket is leached with hot 1 : 1 HNO_3 solution. The leach solution together with any loose solids is transferred to a beaker and digested, with addition of small quantities of concentrated HNO_3 when necessary, until solution is essentially complete. The solution is evaporated to dryness, and the residue is converted to chloride by repeated evaporations with concentrated HCl. Water is

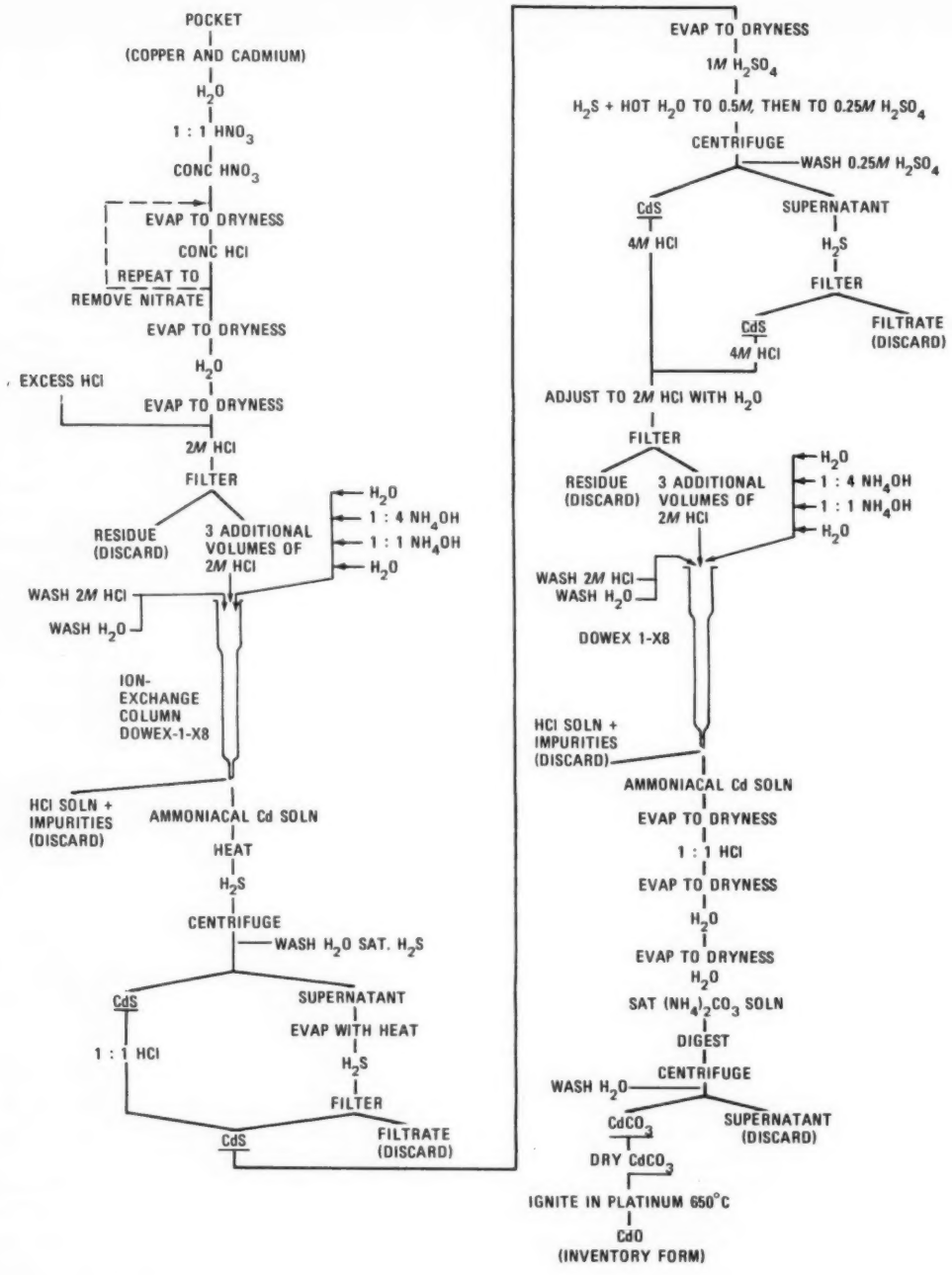

Fig. 1*b* Schematic diagram of the chemical purification procedure for cadmium isotopes collected in copper pockets.

used to dissolve the residue, and the solution is evaporated to dryness to remove excess HCl. After dissolving the residue in 2*M* HCl and filtering, a volume of 2*M* HCl is added which is three times that of the solution.

The 2*M* HCl solution of cadmium is passed through an ion-exchange column of Dowex 1-X8 resin that has been conditioned by passing two (or more) column volumes of 2*M* HCl through the column. After introduction of the cadmium, the column is washed with 2*M* HCl and water, and the washings are discarded. The cadmium is eluted from the column by first washing with two column volumes of water, then with a minimum of two column volumes of 1 : 4 NH_4OH, followed by further washings with 1 : 1 NH_4OH and water. The combined water washes and NH_4OH solution are heated, H_2S is passed through the solution, and the CdS precipitate is allowed to settle overnight. The supernatant liquid is decanted, and the precipitate is centrifuged and washed with water saturated with H_2S. The supernatant liquid is tested for complete precipitation of cadmium. (If cadmium has not been completely precipitated, the supernatant liquid is heated to decrease the volume and treated again with H_2S.) Any CdS at this point is recovered by filtration on Whatman No. 42 paper. The CdS in the centrifuge tube is dissolved in 1 : 1 HCl, and this solution is passed through the filter paper containing any CdS from the supernatant liquid.

The $CdCl_2$ solution is evaporated to dryness and the residue dissolved in 1*M* H_2SO_4. While H_2S is being introduced into this solution after heating, the acidity is reduced by the addition of hot water, first to 0.5*M* and then to 0.25*M* H_2SO_4. (Any zinc impurity does not precipitate readily under these acid conditions.) The CdS is allowed to settle overnight. The mixture is centrifuged, and the combined supernatant and wash solutions of 0.25*M* H_2SO_4 are treated with H_2S. Any CdS recovered is collected on Whatman No. 42 filter paper. The residues in the centrifuge tube and on the filter paper are dissolved in hot 4*M* HCl, combined, and then diluted with water to 2*M* HCl. The solution is again filtered.

The solution is passed again through the ion-exchange column and eluted with 1 : 4 NH_4OH, then 1 : 1 NH_4OH, and washed with water as before. The water and NH_4OH solutions are combined and evaporated to dryness on a steam bath; the residue is now dissolved in 1 : 1 HCl and again evaporated to dryness, and finally it is dissolved in water and evaporated to dryness.

The solid $CdCl_2$ is dissolved in water, and a saturated aqueous $(NH_4)_2CO_3$ solution is added in excess to ensure quantitative precipitation of $CdCO_3$. The mixture is digested on a steam bath and then centrifuged. The supernatant liquid is decanted, and the precipitate is washed with water and then dried in the centrifuge tube. The $CdCO_3$ is transferred to a platinum dish and ignited at 650°C to CdO, the stable inventory form.

Chlorine

Chlorine has two naturally occurring isotopes—^{35}Cl and ^{37}Cl—with abundances of 75.4 and 24.6%, respectively. The pockets used for collecting these isotopes are more complicated than those for the isotopes of most other elements. The pocket itself is constructed of copper and contains both a magnesium-metal reflecting plate and magnesium-metal turnings. Magnesium is used to combine with the (slightly energetic) sputtered chlorine. In the purification and recovery, the various pocket components are treated separately, and, when the chlorine-isotope deposits from the various pocket components have been brought into solution, the several solutions are combined. The chloride is precipitated as AgCl which in turn is heated in hydrogen, and the resulting HCl gas is collected in water and a somewhat less than stoichiometric amount of NaOH. The slight excess of HCl in the combined solutions is titrated with 0.1000*M* NaOH. The NaCl solution is evaporated to dryness, ignited, dissolved in water, filtered, and evaporated to crystals of NaCl, the stable inventory form.

The detailed procedure for the purification of chlorine isotopes is shown in Fig. 2. The copper plate defining the beam slit is first removed from the pocket, and the pocket opening is closed with masking tape to prevent entry of wash solution or loss of loose particles from within the pocket. Only the washings (H_2O and 1.5*M* HNO_3) from the inner side of the plate (against the pocket) are retained.

The outside of the pocket is cleaned with hot 1 : 1 HNO_3, with the washings being discarded. The tape is carefully removed, and the area that was covered by the tape is washed with a small piece of sponge moistened with 1 : 1 HNO_3; entry of the wash solution into the pocket is avoided, and both the washings and the sponge are discarded.

The magnesium turnings are transferred (keeping them dry to prevent evolution of HCl gas) from the pocket to a three-neck, round-bottom, distilling flask with an inlet for nitrogen, one for a dropping funnel,

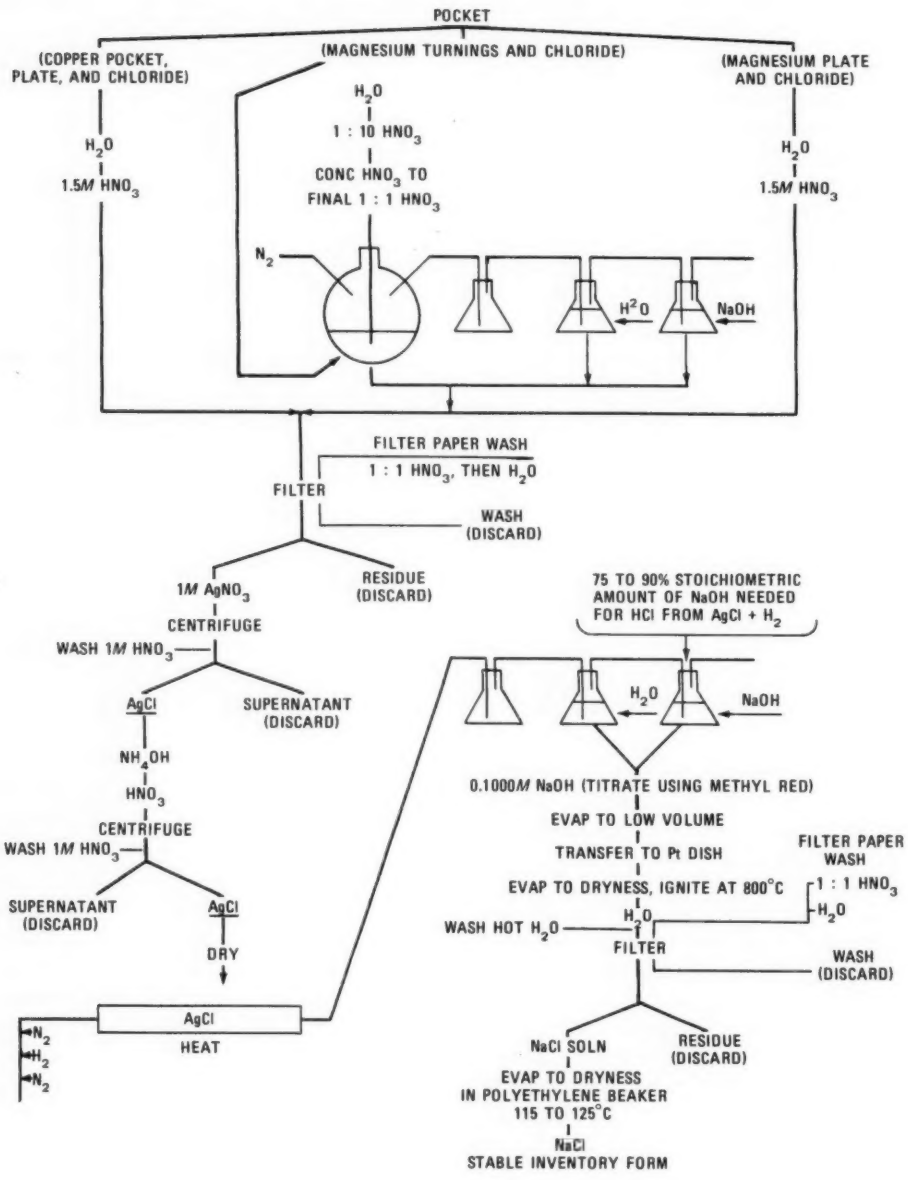

Fig. 2 Schematic diagram of the chemical purification procedure for chlorine isotopes.

and one for connection to a series of gas-washing bottles for collection of the liberated HCl gas. The series contains first an empty bottle as a safety trap, then a gas-washing bottle containing H_2O, and finally one containing NaOH solution. The system is purged with nitrogen gas for 10 min, water is then added slowly from the dropping funnel until the reaction is complete, and finally 1 : 10 HNO_3 is cautiously added. The flask is warmed with a heating mantle until the reaction subsides, and then concentrated HNO_3 is added—dropwise at first—until the concentration of the solution is 1 : 1 HNO_3. Additions of acid are continued until all magnesium turnings are in solution. This solution is combined with those from the gas-washing bottles and reserved.

The magnesium reflecting plate is removed from the pocket, washed with water, and then with 1.5*M* HNO_3 until all the $MgCl_2$ is in solution.

The copper pocket is processed by washing the inside first with H_2O and then with 1.5*M* HNO_3 until the wash solution gives a negative test for Cl^- (absence of AgCl precipitate upon addition of $AgNO_3$ to a test portion of the wash solution).

All the chloride-containing solutions are combined and filtered on a Whatman No. 42 paper that has been washed with 1 : 1 HNO_3 and then with water to remove any traces of Cl^- from the paper. The residue is discarded.

The recovered chlorine isotope is precipitated as AgCl by addition of 1*M* $AgNO_3$, allowed to settle, centrifuged, and washed with 1*M* HNO_3. The AgCl residue is now dissolved in NH_4OH solution and then reprecipitated by addition of HNO_3 solution until the solution is acidic. The precipitate is centrifuged, washed with 1*M* HNO_3, and dried at 110°C.

The precipitate is weighed and then transferred to a porcelain boat that in turn is placed in a quartz-tube, horizontal, electric furnace. As with the distillation flask, glass scrubbing bottles are connected to the discharge of the furnace, using ground-glass connections. The first is used as a trap, the second contains H_2O, and the third contains a quantity of NaOH in solution that is from 70 to 90% of the stoichiometric amount (as determined from the weight of AgCl) required to trap the hydrogen chloride formed in the subsequent reduction of AgCl with H_2.

The system is purged with N_2, and then the flow of H_2 is started. The furnace is heated slowly to 400°C, held at this temperature for ~2 hr, and then heated at 750°C for ~8 hr or until the reduction is complete.

The furnace is cooled with a flow of N_2, and when cool, the scrubbers are removed and the solutions from the scrubbers are combined in a beaker. A 0.1000*M* NaOH solution is used to titrate this slightly acidic solution to a methyl red end point (this prevents the presence of sodium in forms other than NaCl in the final product). The sodium chloride solution is evaporated to a low volume and then transferred to a platinum dish, evaporated to dryness, dried overnight at 125 to 135°C, and then ignited at 800°C for ~45 min. After cooling, the salt is dissolved in water and filtered through a Whatman No. 42 filter paper that has been previously washed to remove traces of chloride. The residue is washed with hot water.

The filtered sodium chloride solution is transferred to a polyethylene beaker, evaporated to near dryness on a steam bath, and dried in an electric oven at 115 to 125°C. The sodium chloride is the stable inventory form.

Indium

Indium has two naturally occurring isotopes—^{113}In and ^{115}In—with abundances of 4.23 and 95.77%, respectively. Indium-115 is very, very, slightly unstable, having a half-life of about 6×10^{14} years and decaying only by beta emission, with a maximum energy of 0.48 MeV.

Indium isotopes are collected in graphite pockets, and the subsequent chemical purification process is relatively simple. Indium in nitrate solution from the leached and ignited pocket is precipitated as $In(OH)_3$ with NH_4OH, followed by dissolution and reprecipitation as In_2S_3, then precipitated again as $In(OH)_3$ and ignited to In_2O_3, the stable inventory form.

The detailed procedure for the purification of indium isotopes is shown schematically in Fig. 3. The graphite collector pocket for each isotope is cleaned in the usual way,[1] i.e., buffing with aluminum oxide-coated abrasive cloth, cutting off the heavy ends of the pocket, removal of the protective tape, and crushing the pocket in a plastic bag. The graphite is transferred to a Soxhlet extractor and extracted overnight with a 1 : 1 HNO_3 solution. The leach solution and water washings from the graphite are decanted and retained, and the graphite is dried and then ignited in oxygen in a quartz boat within a quartz tube in an electric furnace at 700 to 800°C.

The ash from the graphite ignition is dissolved in concentrated HNO_3 and combined with the leach and wash solutions before filtering to remove any insolubles. The filtrate is evaporated to near dryness, and water is added to bring the indium concentration to 0.1 g/200 ml solution.

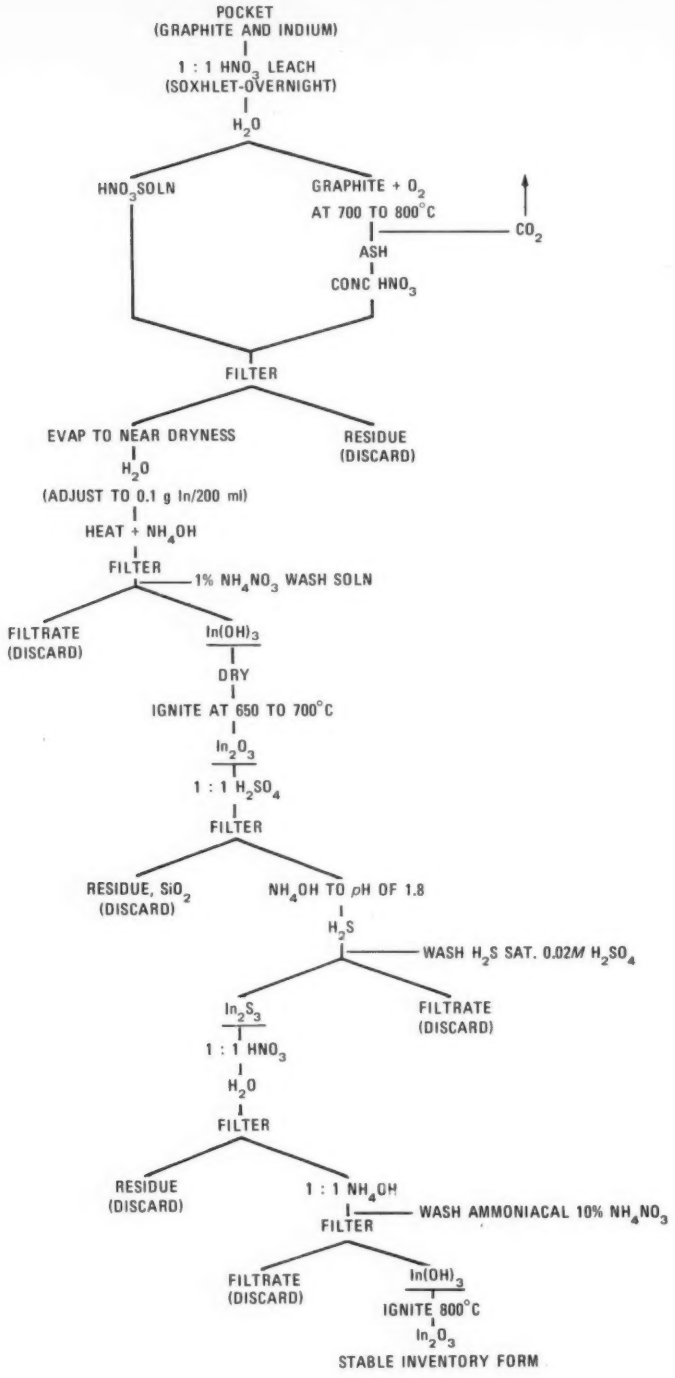

Fig. 3 Schematic diagram of the chemical purification procedure for indium isotopes.

The solution is heated to near boiling, and indium is precipitated as $In(OH)_3$ by the addition of a slight excess of NH_4OH solution. The precipitate is filtered by suction on a Whatman No. 42 filter paper and washed with a slightly ammoniacal 1% NH_4NO_3 solution.

The filter paper and residue are dried and then gradually ignited at 650 to 700°C. (Ignition must be under well-controlled oxidizing conditions with special care exerted to avoid flaming the filter paper which might cause partial reduction of indium to metal and loss by volatilization.)

The In_2O_3 is then dissolved in 1 : 1 H_2SO_4 and filtered to remove silica. The acidity of the filtrate is adjusted to a *p*H of 1.8 with dilute NH_4OH, and the solution is then saturated with H_2S to precipitate In_2S_3. The precipitate is filtered on a sintered-glass frit and washed with 0.02*M* H_2SO_4 saturated with H_2S. The In_2S_3 is dissolved in 1 : 1 HNO_3 and the solution diluted with water and filtered on a Whatman No. 42 paper. A 1 : 1 NH_4OH solution is added to again precipitate $In(OH)_3$; the precipitate is filtered on paper and washed with warm, slightly ammoniacal 10% NH_4NO_3 solution.

The filter paper with the residue is transferred to a platinum dish and carefully dried, charred, and ignited—without flaming—at a maximum temperature of 800°C for several minutes. The In_2O_3 product is the stable inventory form.

Palladium

Palladium has six naturally occurring isotopes with abundances as follows:

Isotope	Natural abundance, at.%	Isotope	Natural abundance, at.%
^{102}Pd	0.96	^{106}Pd	27.33
^{104}Pd	10.97	^{108}Pd	26.71
^{105}Pd	22.23	^{110}Pd	11.81

The chemical purification of palladium isotopes is as follows. The graphite pockets are ignited, and the palladium oxide is reduced to metallic palladium with formic acid, which, in turn, is dissolved in aqua regia for subsequent precipitation with dimethylglyoxime. This precipitate is dissolved in hot 1 : 1 HNO_3 and converted to the sulfate with H_2SO_4, and then the palladium is reprecipitated as the metal, the stable inventory form.

The detailed procedure for the purification of palladium isotopes is shown schematically in Fig. 4. The various isotopes are collected in graphite pockets, and each pocket is cleaned and treated in the usual way[1] and then ignited at 750 to 800°C in oxygen in a quartz boat within a horizontal quartz or Vycor tube in an electric furnace. After the graphite is consumed, the boat is removed from the ignition tube and allowed to cool. The residue is covered with formic acid (HCOOH) to reduce the PdO to palladium metal—conversion to the metallic state permits faster dissolution of the ignited material. The formic acid in the boat is evaporated to dryness at a low temperature.

The residue is transferred with water and hot (if necessary) 1 : 1 HNO_3 into a beaker and evaporated to near dryness before adding ~25 ml of aqua regia to effect dissolution. Successive additions of aqua regia and gentle heating may be required to completely dissolve the palladium.

The solution is evaporated to near dryness, and HNO_3 is driven off by repeated HCl additions and evaporations. Complete removal of HNO_3 is indicated when dropwise additions of HCOOH to the solution yield no evolution of NO_2. The acidity is adjusted to a *p*H of 0.60 by dilution, and the solution is filtered through a Whatman No. 42 paper to remove silica and other insoluble materials.

Palladium is then precipitated from the filtrate at room temperature by the addition of 1.5% dimethylglyoxime/ethyl alcohol solution, and, after settling, the precipitate is filtered on a medium-frit glass funnel and washed, first with 2% HCl, then with water, and finally with ethyl alcohol. [Palladium dimethylglyoxime can be ignited directly to the metal (the preferred inventory form), but volatility loss may occur if the ignition is not performed slowly and carefully. Hence, to ensure that no loss occurs, the palladium dimethylglyoxime is dissolved, and the palladium is subsequently precipitated as the metal.]

The palladium dimethylglyoxime, $Pd(C_4H_7O_2N_2)_2$, is dissolved slowly in the funnel with hot 1 : 1 HNO_3, using suction. When all the precipitate has dissolved, the filtrate is transferred to a beaker and evaporated to dryness. The organic matter is decomposed by boiling with small additions of concentrated HNO_3. After cooling, concentrated H_2SO_4 is added and the solution heated to remove HNO_3. The solution is allowed to cool and then is adjusted to about 0.25*M* H_2SO_4. Silica-free ammonium hydroxide is added until the solution becomes slightly alkaline, and then HCOOH is added until the solution is slightly acid; heating to boiling causes the palladium metal to

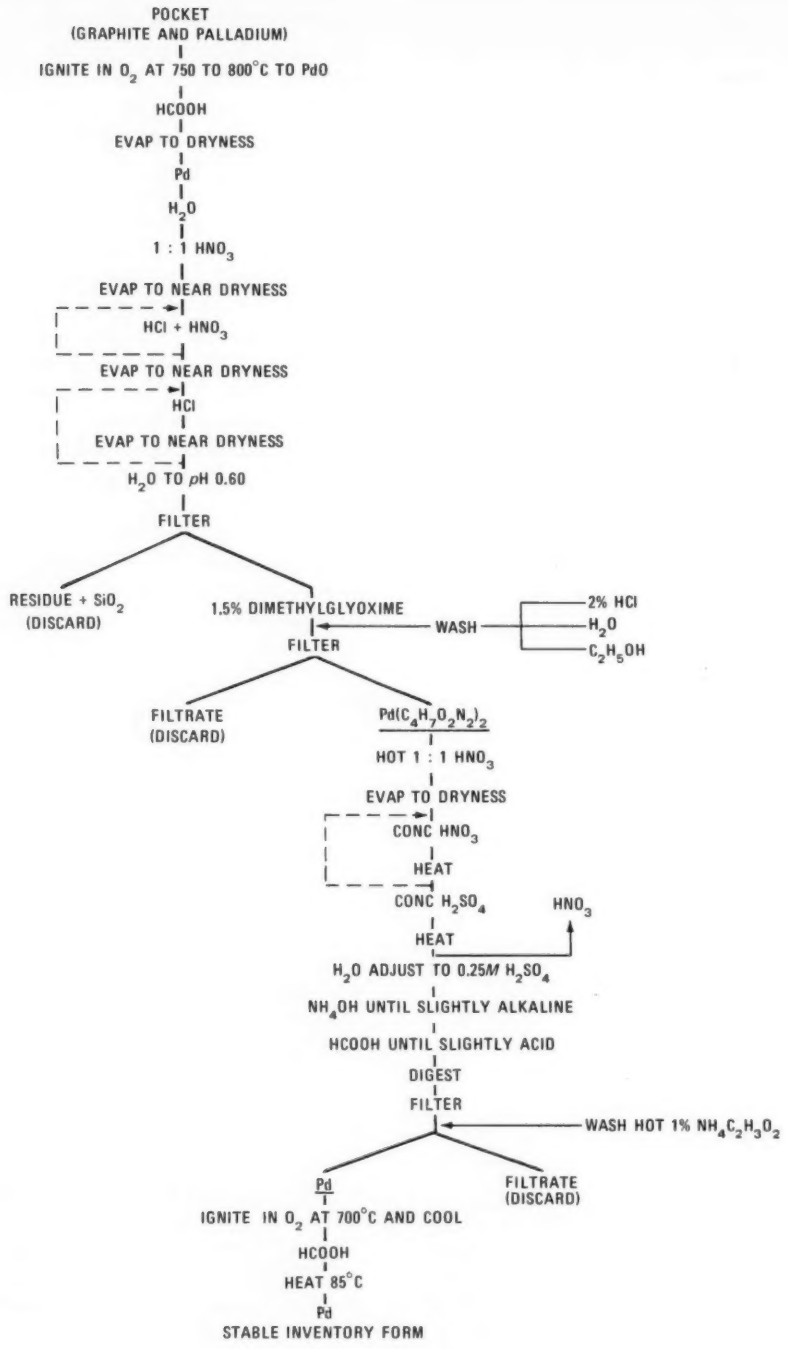

Fig. 4 Schematic diagram of the chemical purification procedure for palladium isotopes.

precipitate and coagulate. The palladium is filtered on a Whatman No. 42 paper and washed with 1% ammonium acetate solution. Palladium adhering to the beaker is removed using a stirring rod and small piece of filter paper; the paper is added to the filter funnel and washed with 1% ammonium acetate.

The filter paper and its contents are transferred to a Vycor crucible and ignited slowly up to 700°C in an oxygen atmosphere to decompose the carbonaceous matter. After cooling, the palladium is moistened with HCOOH and heated at 85°C until dry. The palladium metal is the stable inventory form.

Potassium

Potassium has three naturally occurring isotopes—^{39}K, ^{40}K, and ^{41}K—with abundances of 93.08, 0.012, and 6.91%, respectively. Potassium-40 is very slightly unstable, having a half-life of 1.3×10^9 years and decaying by beta emissions of 1.31-MeV maximum energy, electron capture, and gamma radiation of 1.46 MeV (11%).

The abundances of potassium isotopes differ enough to require the pocket for the ^{39}K isotope, because of its high abundance, to be larger than those for the other isotopes and to contain a graphite striking pad. Because of the extreme reactivity of metallic potassium, special safety precautions[5] must be taken in disassembling the receiver and dissolving the metallic potassium collected in the pockets. Since the amount of ^{39}K collected is considerably greater than that of ^{40}K or ^{41}K, the purification procedure for ^{39}K is more complicated than that for the other two isotopes. Copper from the leach solution is removed by electrolysis, a portion of other impurities are removed as insoluble sulfides, sulfur as sulfate is precipitated as $BaSO_4$, and silica is dehydrated with perchloric acid and removed by filtration. After the removal of silica, the potassium isotope is recovered by precipitation from an alcoholic perchlorate solution as $KClO_4$ and subsequently ignited and decomposed to KCl, the stable inventory form. In the purification of ^{40}K and ^{41}K, the sulfide and sulfate precipitations are replaced by a separation of impurities by electrolysis with a mercury cathode; the purified isotopes are recovered as insoluble perchlorates, as with ^{39}K, and ignited to KCl.

Purification of ^{39}K. The detailed procedure for the purification of the ^{39}K isotope is shown schematically in Fig. 5*a*. The ^{39}K is collected in a large copper pocket containing a graphite striking pad. The outside of the copper pocket is cleaned in the usual way[1] with water and 1 : 1 HNO_3, and the protective tape is taken off. The graphite striking pad is removed from the pocket (behind a suitable safety shield) and transferred to a beaker and leached first with water and then with 1 : 1 HNO_3.

The graphite pad is then broken into small pieces, transferred to a large Soxhlet extractor, and extracted for about 24 hr with 1 : 1 HNO_3. All the leach solutions from the graphite pad are retained.

The copper pocket is placed in a large Pyrex tray (again behind a suitable safety shield), and the inside of the pocket is flushed with water until essentially all the potassium is in solution. This solution is transferred to a beaker, and the pocket is treated with 1 : 1 HNO_3 to remove the black deposits on the copper. When the copper metal remains bright after addition of 1 : 1 HNO_3, all the potassium is assumed to be in solution.

All the leach solutions are combined (the solution should be acid; if not, HNO_3 is added until the solution is acidic), and the solution is digested at a low heat. Insolubles are removed by filtering the solution through a Whatman No. 42 paper, using a platinum filter cone and gentle suction.

The filtrate is evaporated to dryness on a steam bath to remove excess HNO_3, and the residual solid is dissolved in water and the minimum amount of HNO_3 necessary to effect solution. This solution is electrolyzed, using platinum electrodes and about 1 A of current until all the copper and lead have been removed. The solution is again evaporated to dryness and the nitrate decomposed by repeated evaporations with concentrated HCl and finally with concentrated HCl and formic acid (HCOOH).

The resulting solution is diluted with water, saturated with H_2S, digested at near boiling, and filtered; the filtrate is made alkaline with silica-free NH_4OH, and the solution is again saturated with H_2S, digested, and filtered. The residue is washed with 1 : 10 $(NH_4)_2S$ and discarded.

Concentrated HCl is used to acidify the filtrate, which is boiled to liberate H_2S and to coagulate any elemental sulfur. The solution is filtered on a Whatman No. 42 paper, evaporated to near dryness, and treated with aqua regia to eliminate the ammonium salts. Water and a few milliliters of dilute $BaCl_2$ solution are added to precipitate any sulfate formed by oxidation of sulfur, and the solution is filtered to remove $BaSO_4$.

Perchloric acid is added to the filtrate, and the solution is evaporated to fumes of $HClO_4$. After cooling, the salts are dissolved in water, and the dehydrated silica is removed by filtering. The addition of $HClO_4$ and the evaporation to fumes are repeated.

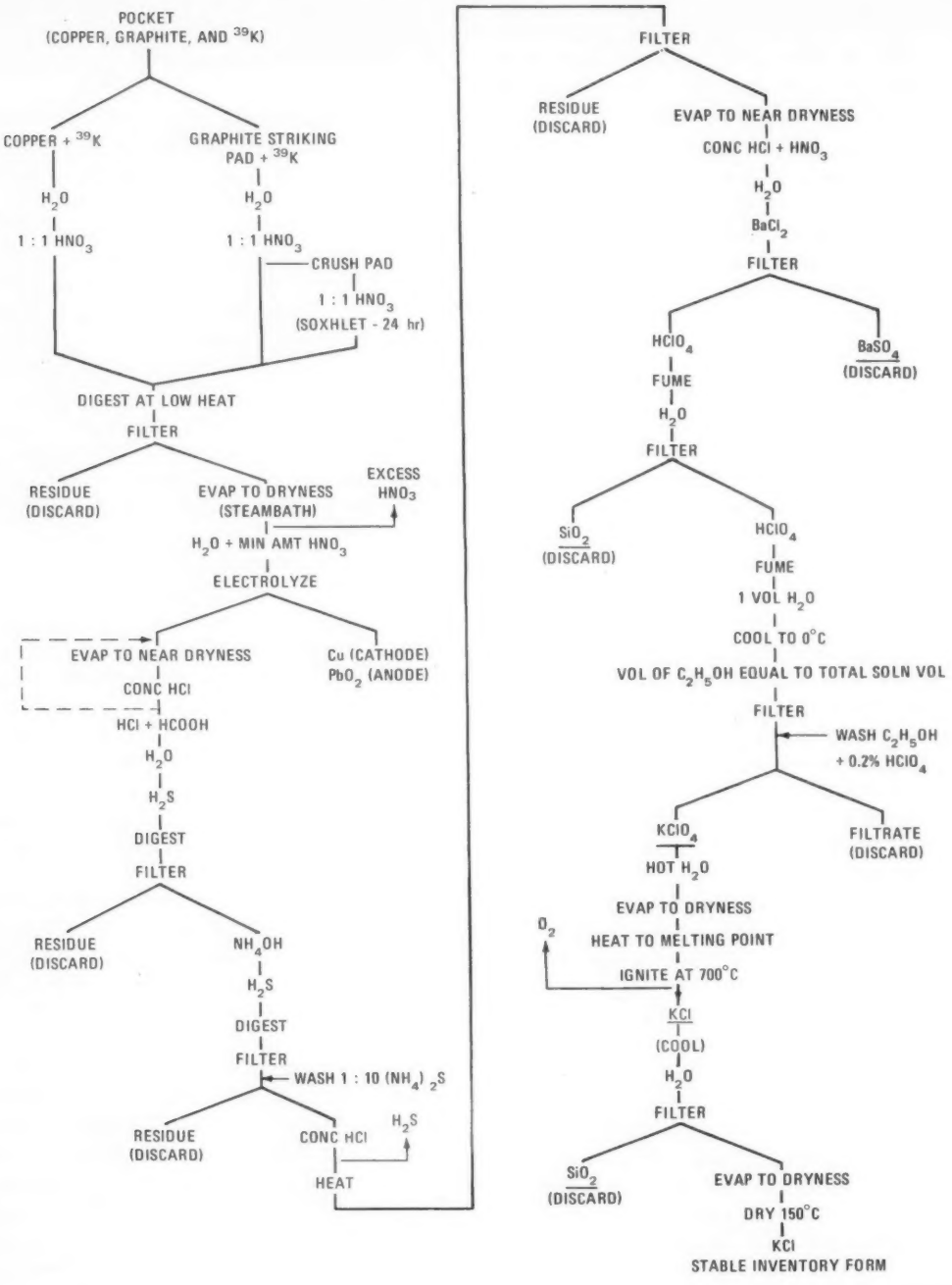

Fig. 5*a* Schematic diagram of the chemical purification procedure for the ^{39}K isotope.

The perchloric acid solution is diluted with an equal volume of water and cooled to 0°C. A volume of absolute alcohol equal to the total volume of solution is then added to the perchloric acid solution to precipitate $KClO_4$, which is allowed to settle for about 15 min and then recovered by filtering the solution with a medium frit glass funnel. Cold alcohol with 0.2% $HClO_4$ is used for washing the $KClO_4$.

The $KClO_4$ is dissolved through the frit with hot water and transferred to a Vycor dish. The solution is evaporated to dryness and the residue heated slowly to the melting point of $KClO_4$. After most of the bubbling has ceased, the temperature is increased to 700°C to ensure complete decomposition to KCl. (A loose-fitting platinum cover helps to prevent spattering losses.) The residue is cooled and dissolved in a minimum amount of water; the resultant solution is filtered through a Whatman No. 42 paper to remove silica. The filtrate is evaporated to dryness, and then the KCl residue, the stable inventory form, is dried in an oven at 150°C.

Purification of ^{40}K and ^{41}K. The detailed procedure for the purification of ^{40}K and ^{41}K is shown schematically in Fig. 5*b*. The copper pocket is cleaned on the outside in the usual way,[1] and then the potassium deposit is dissolved from the inner surfaces of the pocket with water, followed by leaching with 1 : 1 HNO_3 until the surface of the copper remains bright. The water and nitric acid solutions are combined, digested until any copper flakes and particles have dissolved, and filtered on a Whatman No. 42 paper. The filtrate is evaporated to dryness on a steam bath to remove excess HNO_3; the residue is dissolved in water and a minimum amount of HNO_3, and the solution is electrolyzed with platinum electrodes until copper and lead have been removed.

After removal of the electrodes, 10 to 15 ml of $HClO_4$ is added, and the solution is evaporated to dryness. The salts are dissolved in water, 2 ml of $HClO_4$ is added, and the solution is electrolyzed, using a mercury cathode and platinum anode at about 0.15 A/cm^2.

After electrolysis the solution is transferred first to a beaker and evaporated to a low volume and then to a small quartz container and evaporated to dryness. The residue, $KClO_4$, is heated to 700°C; heating is discontinued when all $KClO_4$ has decomposed to KCl. The KCl is dissolved in a minimum amount of hot water and filtered through a Whatman No. 42 paper; the KCl solution is evaporated again to a low volume, transferred to a small, weighed vial, and evaporated to dryness. The vial with the KCl is dried in an oven at 150°C. The KCl is the stable inventory form.

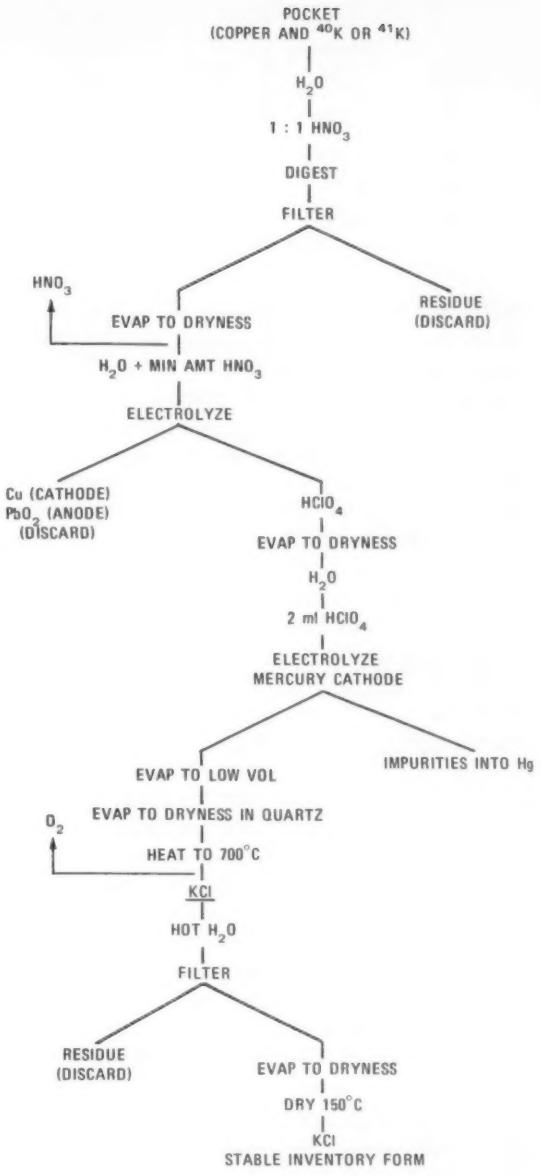

Fig. 5*b* **Schematic diagram of the chemical purification procedure for ^{40}K or ^{41}K isotopes.**

Ruthenium

Ruthenium has seven naturally occurring isotopes with abundances as follows:

Isotope	Natural abundance, at.%	Isotope	Natural abundance, at.%
^{96}Ru	5.47	^{101}Ru	17.10
^{98}Ru	1.84	^{102}Ru	31.70
^{99}Ru	12.77	^{104}Ru	18.56
^{100}Ru	12.56		

Ruthenium isotopes are collected in graphite pockets which are then ignited in oxygen. Both the volatile RuO_4 and the ruthenium residue (ash) are recovered. The ash is fused with a $KOH–KNO_3$ mixture to bring the ruthenium into solution. Ruthenium from the combined ash and trap solutions is again volatilized, as RuO_4, and collected in HCl solution saturated with SO_2. Ruthenium is precipitated as the sulfide, ignited to the oxide, and reduced to the metal, the stable inventory form, by ignition in hydrogen.

The detailed procedure for the purification of ruthenium isotopes is shown schematically in Fig. 6. The graphite pockets are cleaned on the outside in the usual way[1] and then ignited in oxygen at 650°C in a silica boat in a Vycor tube in an electric furnace. Volatile RuO_4, formed during the ignition, is collected in 1 : 1 HCl containing SO_2. The residue from the graphite ignition is fused with a mixture of 5 parts KOH : 1 part KNO_3 in a silver container at ~600°C. After cooling, the melt is dissolved in water in a plastic vessel, and, after the silver container has been removed, the solution is made strongly acid with concentrated HCl. The solution is digested in a steam bath and diluted by adding the solution from the trap used during ignition of the graphite. The combined solutions are filtered on a Whatman No. 42 paper, the residue is washed with hot H_2O to remove Cl^-, and the filtrate is reserved.

The filter paper is ignited, and the residue is fused with a small quantity of 5 KOH : KNO_3 as before; the melt is dissolved in water in a plastic vessel, and the solution is acidified with HCl and filtered into the reserved solution.

The resultant solution is evaporated to near dryness, about 10 ml of concentrated H_2SO_4 is added, and the mixture is evaporated to heavy fumes of SO_3. After cooling, about 100 ml of water is carefully added, and the slurry is transferred to a three-neck distillation flask. The distillation apparatus is all glass, and the ground-glass joints are lubricated with concentrated H_2SO_4. One neck of the flask contains a passage for filtered air to be drawn through the system; the center neck contains an open top, cylindrical, separatory funnel; and the third neck contains a delivery tube to a series of three receiving traps. All the traps contain 1 : 1 HCl saturated with SO_2. The first and second traps are provided with a separatory funnel for additions of 6% H_2SO_3 as the distillation proceeds.

About 100 ml of filtered 10% $NaBrO_3$ solution is also added to the flask, and the solution is heated gently to boiling while filtered air is drawn through the system. During the distillation of RuO_4, 6% H_2SO_3 is added to the first and second traps occasionally as the distillation proceeds.

After the ruthenium has been distilled, the trap solutions are transferred to a single beaker (under a fume hood) and evaporated to a moist residue on a steam bath. Enough 1 : 1 HCl is added to dissolve the residue, and the solution is then diluted with water and filtered to remove silica.

The filtrate is evaporated to near dryness on a steam bath, ~10 ml of H_2SO_4 is added, and the solution is heated until all the HCl has been volatilized. The solution is cooled, diluted, and filtered, and the residue is washed with 1 : 20 H_2SO_4. The residue is reserved. The filtrate is saturated with H_2S for about 3 hr, the precipitate is allowed to settle until the supernatant liquid is clear (overnight), and then the contents of the beaker are digested.

The ruthenium sulfide is filtered on paper and washed with a 2% $(NH_4)_2SO_4$ solution saturated with H_2S. The filter paper previously reserved from the filtration of sulfate solution and the paper containing the RuS_2 are both placed in a single Vycor crucible and covered with crystalline $(NH_4)_2SO_4$ to prevent deflagration of the sulfide during the subsequent ignition. The paper is dried slowly, and then the paper and the residue are ignited at 700°C.

The residue is transferred to a Vycor boat that is placed in a horizontal quartz tube in an electric furnace. The ruthenium oxide is reduced to ruthenium metal by ignition up to 800°C under dry hydrogen; the hydrogen flow is continued during the cooling period. Ruthenium metal is the stable inventory form.

Tantalum

Tantalum has only two naturally occurring isotopes—^{180}Ta and ^{181}Ta—and their ratio of abundances is ~1 : 10,000 (i.e., 0.012 and 99.998%, respectively).

Because of the very high natural abundance of ^{181}Ta, only the less abundant isotope ^{180}Ta is

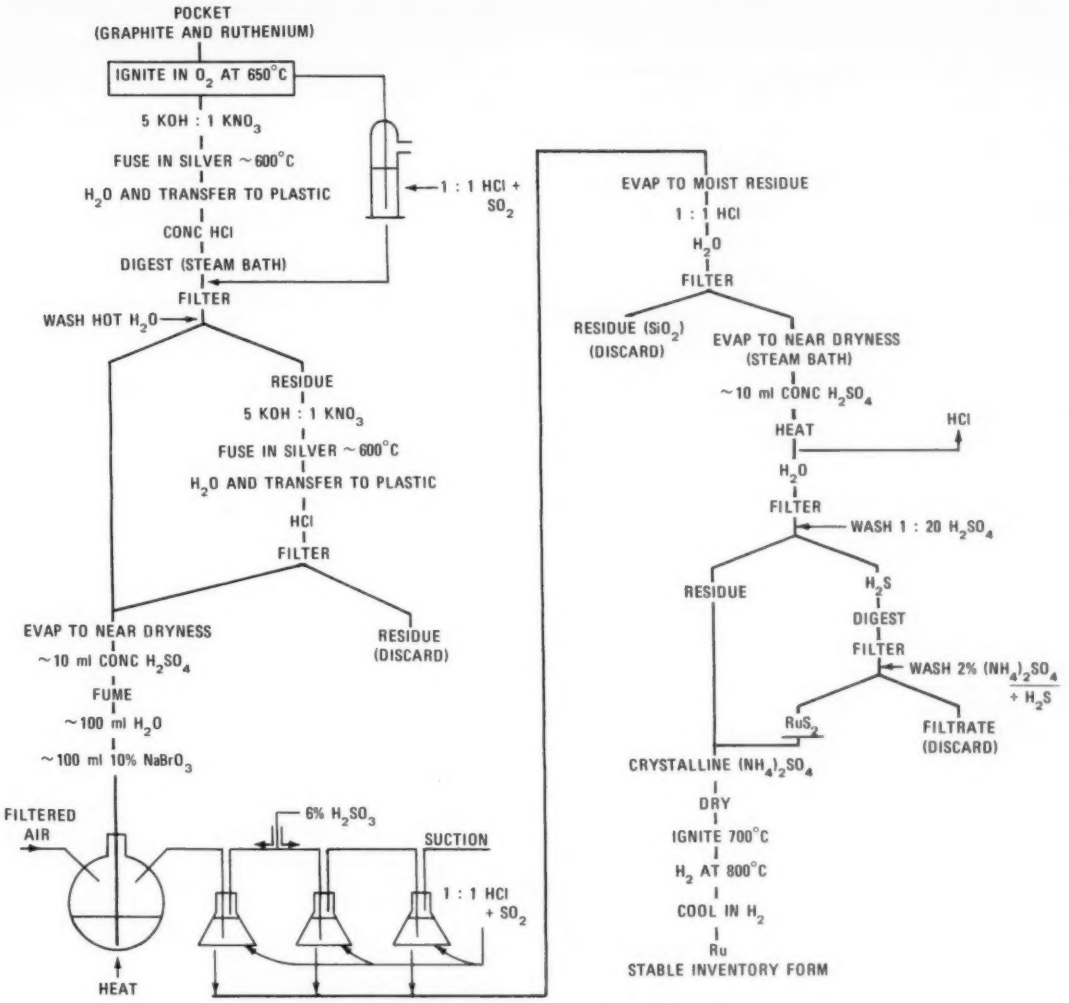

Fig. 6 Schematic diagram of the chemical purification procedure for ruthenium isotopes.

chemically purified. In general, the purification procedure is relatively simple. Tantalum is brought into solution with a mixture of nitric and hydrofluoric acids, repeatedly precipitated as $Ta(OH)_5$ and then precipitated as Ta_2O_5 from fuming perchloric acid.

The detailed procedure for the purification of tantalum isotopes is shown schematically in Fig. 7. The graphite pocket containing the ^{180}Ta isotope is cleaned on the outside in the usual way,[1] and then any dust dislodged during the scraping loose of the deposit in the interior of the pocket is collected with a vacuum attachment that allows depositing the material on a sintered-glass funnel. The collected loose material is ignited in a platinum dish in an oxygen atmosphere at 600°C. (The graphite pocket is not ignited.) The residue after ignition, impure Ta_2O_5, is dissolved by heating in the presence of repeated additions of a mixture of 1 part concentrated HNO_3 : 4 parts HF. The solution is diluted with water and then filtered on paper through a plastic funnel into another platinum dish. Tantalum is precipitated as $Ta(OH)_5$ by the addition of NH_4OH. The precipitate is filtered, dissolved with 1 : 1 HCl that is ~1*M* in HF, reprecipitated with NH_4OH, and filtered. The dissolution, precipitation, and filtration steps are repeated until there is no evidence of traces of copper (small quantities of copper

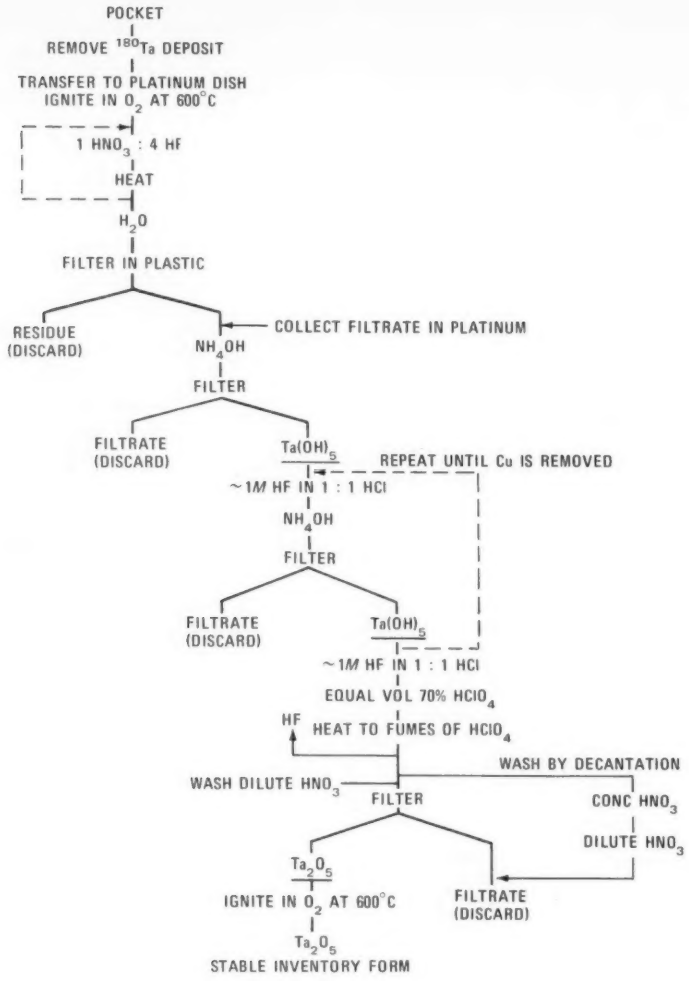

Fig. 7 Schematic diagram of the chemical purification procedure for the ^{180}Ta isotope.

are sputtered from the internal parts of the calutron into the pocket).

The copper-free $Ta(OH)_5$ is dissolved in 1 : 1 HCl that is ~1*M* in HF, and then a volume of 70% $HClO_4$ equal to that of the tantalum solution is added. This solution is heated until the heavy fumes of $HClO_4$ are given off (tantalum is held in solution by HF; when the HF is volatilized by the fuming $HClO_4$, quantitative precipitation of tantalum as Ta_2O_5 occurs).

The precipitate is allowed to settle, washed with concentrated HNO_3, and then dilute HNO_3 by decantation. The residue is then collected on a Whatman No. 42 paper and again washed with dilute HNO_3. The paper and residue are transferred to a small platinum dish and ignited at 600°C in oxygen in a horizontal Vycor tube in an electric furnace. The Ta_2O_5 product is the stable inventory form.

Titanium

Titanium has five naturally occurring isotopes as follows:

Isotope	Natural abundance, at.%	Isotope	Natural abundance, at.%
^{46}Ti	7.95	^{49}Ti	5.51
^{47}Ti	7.75	^{50}Ti	5.34
^{48}Ti	73.45		

Titanium isotopes are collected in graphite pockets which are ignited in oxygen. Titanium is precipitated from sulfuric acid solution as metatitanic acid that is separated by filtration and ignited to TiO_2, the stable inventory form.

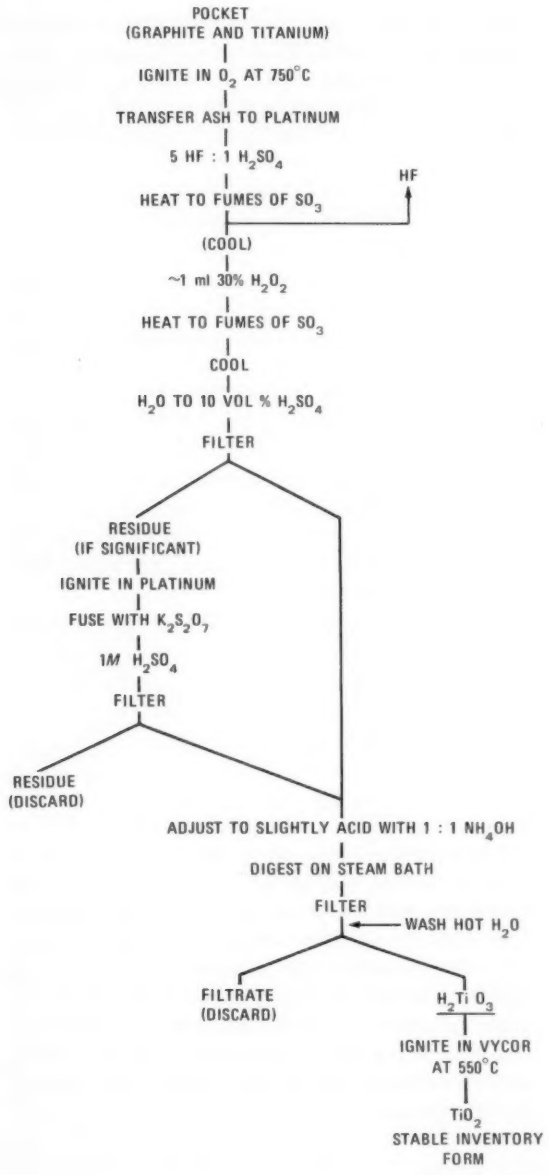

Fig. 8 Schematic diagram of the chemical purification procedure for titanium isotopes.

The detailed procedure for the purification of titanium isotopes is shown schematically in Fig. 8. The graphite pocket for each titanium isotope is cleaned in the usual way,[1] the heavy ends are sawed off, the pocket is crushed and then ignited in a quartz boat in oxygen at 750 to 800°C. The ash is transferred to a large platinum dish and dissolved in a mixture of 5 parts concentrated HF : 1 part concentrated H_2SO_4. [Titanium oxide reacts with HF to form the soluble hexafluorotitanate(IV) ion, TiF_6^{-2}.] The solution is heated until fumes of SO_3 are evolved (an excess of H_2SO_4 must be present to avoid volatilization of TiF_4); the solution is cooled, about 1 ml of 30% H_2O_2 is added, and the solution is again evaporated to fumes of SO_3. This treatment volatilizes the HF and permits the use of glass equipment in the subsequent steps of purification.

The solution is again cooled, carefully diluted with water to bring the acid concentration to 10 vol.% H_2SO_4, and filtered. The filtrate is reserved. If there is any appreciable amount of titanium-bearing residue remaining, it is ignited in platinum, fused with 10 times its amount of $K_2S_2O_7$, dissolved in 1*M* H_2SO_4, and filtered into the reserved filtrate.

The sulfuric acid filtrate is adjusted with 1 : 1 NH_4OH to be just slightly acid and then is digested on a steam bath until metatitanic acid precipitates. The precipitate is recovered by filtration, and the paper and residue are washed with hot water. The paper with the residue is transferred to a Vycor dish for careful drying and subsequent ignition at 550°C for conversion of the metatitanic acid to TiO_2, the stable inventory form.

(REG)

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Xenon-128 Production Process Development*

By D. W. Haase

(Summarized by R. E. Greene)

Supplementary Keywords: *production, stable isotope; production, reactor; volatilization;* ^{126}I; ^{128}I; ^{127}Xe; ^{129m}Xe.

Abstract: *A process for producing the stable isotope* ^{128}Xe *has been developed and demonstrated by Douglas United Nuclear, Inc. Neutron irradiation of elemental iodine produces* ^{128}Xe, *which is separated and recovered in a vacuum system by cryogenic techniques. Approximately 212 ml (STP) of 99.8%* ^{128}Xe *was produced from 450 g of iodine irradiated to a neutron fluence of* 5.4×10^{20} n/cm^2. *This article is a summary of USAEC Report DUN-SA-145.*

Douglas United Nuclear, Inc. (DUN), in response to a solicitation by Argonne National Laboratory (ANL), has developed a method for the production of ^{128}Xe that can be used to provide additional unique tag mixtures in a method for identifying fuel failures in the EBR-II reactor. Previously, enriched stable ^{124}Xe mixed with natural xenon was used as a tag that was introduced into the gas bond of a fuel element during assembly. In the event of fuel failure during irradiation, some of the tagged xenon mixture escapes to the reactor cover gas and can be collected and analyzed, thus providing identification of the failed fuel assembly. The use of ^{128}Xe will increase the flexibility of the tag method.

Development work by DUN, results obtained in the production process for ^{128}Xe, and proving of the process with a small-scale production run are described. Development included design of a target capsule, design and assembly of a gas-separation system, determination of the effects of impurities on product quality, and demonstration of separation and gas-handling techniques. A 160-ml (STP) quantity of ^{128}Xe has been produced, recovered, purified, and shipped to ANL.

EXPERIMENTAL

The only known documentation[1] of ^{128}Xe production in the United States recorded that 0.15 ml (STP) had been produced by Brookhaven National Laboratory by neutron irradiation of potassium iodide. In the present method, elemental iodine was irradiated in the test facilities of the DUN-operated production reactors to produce ^{128}Xe by the nuclear reaction

$$^{127}I + n \rightarrow {}^{128}I \xrightarrow[T_{1/2} = 25\ \text{min}]{\beta^-} {}^{128}Xe$$

Elemental iodine was chosen as the target material because its postirradiation activity levels are low, it has the highest target atom density of all iodine-containing materials, and it can be readily vaporized, thus simplifying the ^{128}Xe separation process.

Iodine for the experimental run was encapsulated in a quartz ampule, which in turn was canned in a high-purity aluminum container to minimize the possibility of product loss. Quartz and aluminum were used for capsule components to minimize postirradiation radioactivity and thus permit unshielded handling and separation operations.

Following irradiation, the ^{128}Xe was extracted from the target capsule in a vacuum separation system consisting of a capsule penetrator vessel for liberating capsule contents into the system, a Dry Ice–cooled trap for iodine removal, a liquid-nitrogen-cooled trap for xenon recovery, and a sample tap for obtaining samples and loading shipping containers. Vacuum pumps capable of evacuating the system to 10^{-3} torr and low-pressure gages were also included. The irradiated capsule was placed in the system, and the system was then evacuated and the capsule punctured. The penetrator vessel was heated to release the xenon from the iodine, and the resulting iodine–xenon gas mixture was passed through the iodine trap to condense the iodine. The remaining xenon migrated to the xenon trap and froze. Impurities with low freezing points—nitrogen, oxygen, and helium—were pumped from the system.

Pilot Capsule

The design of the pilot capsule was based on producing a minimum of 150 ml (STP) of ^{128}Xe from 450 g of iodine and on minimizing the postirradiation

*Summary of Xenon-128 Production Process Development, USAEC Report DUN-SA-145, Douglas United Nuclear, Inc., P. O. Box 490, Richland, Wash. 99352. (Presented at the American Nuclear Society 1970 Winter Meeting, Nov. 15–19, 1970, Washington, D. C.)

radioactivity level to eliminate the need for a hot cell or a shielded separation facility. Quartz was selected for the capsule because it meets the low-radioactivity criterion, can withstand irradiation to high-neutron fluences, and is compatible with iodine at elevated temperatures. A 1.5-in.-diameter by 11-in.-long quartz capsule was loaded with crystalline iodine to 50% theoretical density, evacuated to less than 1 torr, and sealed. The quartz capsule was encased in a thin-wall aluminum container to reduce the potential loss of ^{128}Xe in the event of quartz-capsule breakage during postirradiation handling. The aluminum container was purged with helium prior to sealing to remove air and provide better heat transfer. This quartz–aluminum assembly was housed in a thick-wall, perforated aluminum container for further protection against mechanical damage.

Xenon-Separation System

The stainless-steel system used to recover the product xenon gas (Fig. 1) was designed to separate as much as 1 liter (STP) of ^{128}Xe gas per run. The system includes a capsule penetrator vessel (Fig. 2) that is equipped with an O-ring–sealed puncture pin to liberate the ^{128}Xe gas. A liquid-nitrogen cold trap between the vacuum pump and the penetrator vessel prevents loss of xenon in the event of a capsule leak. A trap for iodine, between the penetrator vessel and the xenon collection system, is large enough to hold all the iodine (4000 g) contained in a production capsule and is cooled with an acetone–Dry Ice mixture. Iodine contamination beyond the iodine trap is negligible because of the low vapor pressure of iodine at Dry Ice temperature.

The xenon collection and transfer portion of the system consists of a xenon trap, a sample port for obtaining both analytical samples and product quantities of ^{128}Xe, a pressure gage calibrated from 0 to 720 in. H_2O, and a helium leak detector that is also used to evacuate the system. During a separation run the xenon trap is cooled with liquid nitrogen (–195°C) to freeze out the xenon, which freezes at –112°C. Since the vapor pressure of xenon at –195°C is 2×10^{-3} torr, gaseous impurities, such as helium or air, that enter the trap can be pumped off with negligible loss of xenon. Xenon is transferred from the trap to the shipping container by warming the trap and cooling the container with liquid nitrogen. To prevent loss of xenon if a leak develops in the system, the xenon trap is sized to permit collection of 1 liter of xenon without exceeding atmospheric pressure.

The helium leak detector is equipped with a low-capacity roughing pump and a diffusion pump and is used to evacuate the xenon-collection portion of the system. Because of the large relative volume of the xenon trap and the small-diameter connecting tubing, the minimum collection-system pressure attainable is 10^{-2} torr. Calculations based on the assumption that the xenon-trap atmosphere, after freezing out of the xenon, is entirely air at 0.1 torr show that the product xenon would have less than 0.5% impurity based on recovery of 100 ml of ^{128}Xe. The total volume of xenon remaining in the 7600-ml system at 0.1 torr would be about 1 ml (STP). Therefore a minimum attainable operating pressure of 10^{-2} torr is satisfactory.

Prior to making a pilot production run, several demonstration runs were made using natural xenon to study xenon handling, collection, and impurity removal. The results predicted a product recovery of 99% and purity of 100%. No iodine was detectable beyond the iodine trap.

Pilot Production Run

Target Irradiation. In the pilot production run, 450 g of iodine was encapsulated in quartz and aluminum, and the capsule was leak checked and irradiated in a test facility of a production reactor to a total neutron fluence of 5.4×10^{20} n/cm^2. At discharge the initial radiation level of the capsule assembly was 200 R/hr at 1 ft in air. Three weeks later the perforated outer can was removed, and the radiation level was 5 R/hr at 2.5 in. in air. When the capsule was placed in the penetrator vessel, a 2-in. thickness of lead-brick shielding decreased the radiation level to less than 10 mR/hr.

Xenon Separation. The penetrator-vessel portion of the system was evacuated through the liquid-nitrogen cold trap to remove helium and air inleakage, primarily from around the penetration pin, and to collect any ^{128}Xe that might have escaped into the aluminum can. After evacuation of the entire system, the penetration pin was driven farther into the penetrator vessel to ensure rupture of the quartz container. The penetrator vessel was heated to approximately 100°C for 2 days to hasten the separation and recovery of the xenon. Periodically the amount of xenon recovered was determined by isolating the xenon cold trap, allowing the trap to warm, and measuring the xenon pressure.

Carbon dioxide inleakage from the air and/or from the Dry Ice trap was removed by introducing 2*M* NaOH solution into the xenon trap and mixing the gas

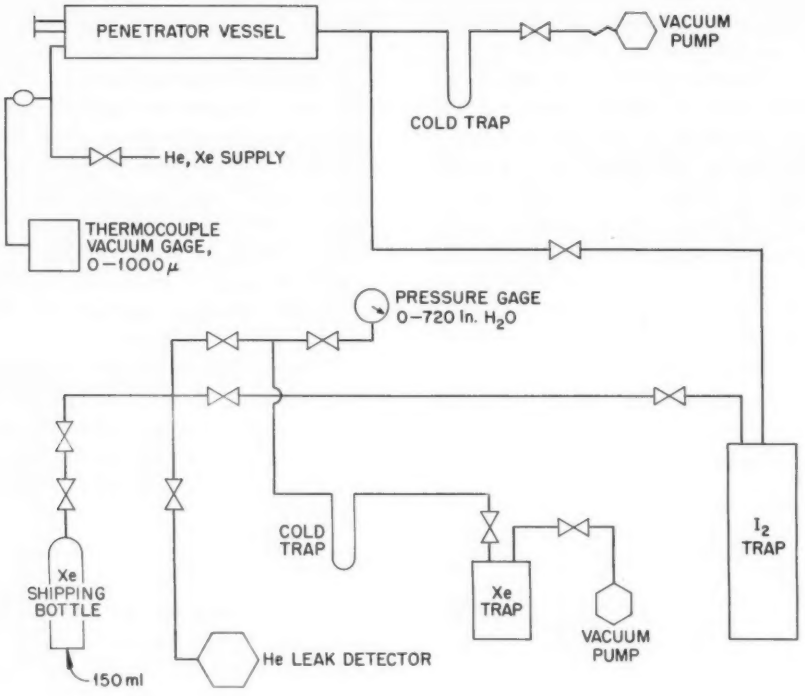

Fig. 1 Xenon separation and collection system.

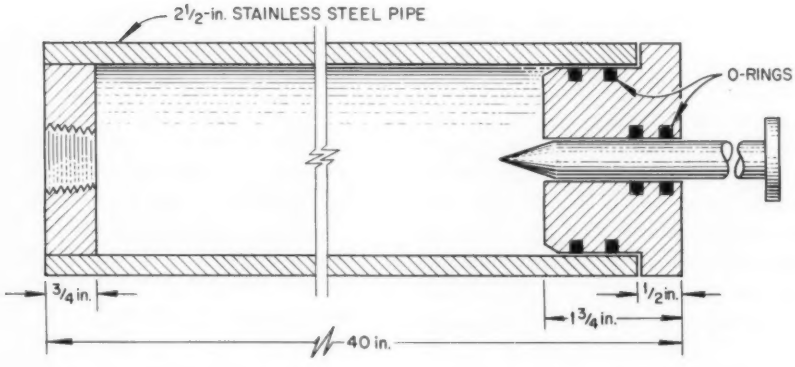

Fig. 2 Capsule penetrator vessel.

and solution. The xenon trap was cooled in ice water to reduce the vapor pressure of H_2O, and the xenon was recovered in a 150-ml shipping bottle cooled with liquid nitrogen. A methanol–Dry Ice cold trap between the xenon trap and the shipping bottle prevented water vapor from entering the shipping container.

Evaluation of the Pilot Run. A volume of 212 ml of ^{128}Xe was recovered from the 450 g of irradiated iodine—a quantity within the limits of uncertainty of the calculated value. The composition of the product, determined by mass spectrometric analysis, was

Gas	Mole %	Gas	Mole %
^{128}Xe	99.8	N_2	0.09
^{129}Xe	0.05	CO	<0.10
CO_2	0.05	He	<0.01
Ar	<0.01	H_2	<0.01
O_2	<0.01	CH_4	<0.01

The 8-day-half-life ^{129m}Xe, produced by burnout of ^{128}Xe, gave a surface reading of 150 mR on the 150-ml sample bottle. (Calculations for both production and burnout of ^{128}Xe are given as appendixes in the full report.) The ^{129m}Xe concentration can be minimized by reducing the total neutron exposure; i.e., for producing a given quantity of ^{128}Xe, a greater quantity of iodine may be irradiated for a shorter length of time.

Also present, as revealed by gamma spectrometry, was a trace amount of ^{127}Xe ($T_{1/2}$ = 36.4 days). The estimated concentration of ^{127}Xe at capsule discharge was 10^{-6} mole %. The presence of this isotope is probably due to a $^{127}I(n,2n)^{126}I$ reaction with 10-MeV neutrons, with approximately 40% of the ^{126}I ($T_{1/2}$ = 12.8 days) decaying to stable ^{126}Xe, which in turn produces ^{127}Xe by neutron capture.

Production Run

A production irradiation capsule of stainless steel featuring both external and internal cooling and capable of containing 4000 g of iodine is now being irradiated for subsequent production of 1500 ml of ^{128}Xe.

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THE STABLE ISOTOPES

By Gheorghe Vasaru, French translation (from Rumanian; Institute of Atomic Physics, Cluj) by Alice Courtot, French Report CEA-Bib-136, 1970.

This bibliographic review of the literature to 1966 was not widely distributed to the western world until the issuance of the French translation in September 1970. Separation methods, analytical determinations, and applications of stable isotopes are described. The report of over 300 pages contains about 950 references. (REG)

Evaluation of Nuclear Reactions That Produce ^{123}I in the Cyclotron

By V. J. Sodd,* K. L. Scholz,* J. W. Blue,† and H. N. Wellman†

Supplementary Keywords: *comparison; production, radioisotope.*

Abstract: *Various nuclear reactions and methods for producing ^{123}I are evaluated and compared.*

Iodine-123 is said to be the most nearly ideal radioisotope of iodine for low-exposure, in vivo measurement.[1] Its use in a human thyroid-uptake test results in the patient's receiving[2,3] a radiation dose only a few percent of that received from the more commonly used ^{125}I and ^{131}I because ^{123}I emits far less charged-particle radiation than does ^{131}I and its physical half-life is only 0.6 and 6.9%, respectively, of that of ^{125}I and ^{131}I. However, preliminary clinical results of the use of ^{123}I for in vivo thyroid measurements suggest that radioiodine contaminants in the product prevent full use of ^{123}I as a high-resolution low-exposure scanning agent.[4,5]

Most of the ^{123}I used in nuclear medical applications has been produced by 15-MeV proton bombardment[6] of enriched ^{123}Te—$^{123}Te(p,n)^{123}I$—or by ^{4}He bombardment[7] of natural antimony—$^{121}Sb(^{4}He,2n)^{123}I$. Both reactions give ^{123}I in good yield, but both products contain longer lived iodine isotopes. The highest purity ^{123}Te obtainable without extraordinary target enrichment is about 79%, and the ^{124}Te, ^{125}Te, and ^{126}Te present give 0.9, 0.08, and 0.3% ^{124}I, ^{125}I, and ^{126}I, respectively[6b] (the ^{130}I content from ^{130}Te is not reported). Contaminants in the product from the natural-antimony target are due to the 42.8% ^{123}Sb present and competing $(^{4}He,n)$ and $(^{4}He,3n)$ reactions. It has been possible[8] to keep the contaminating ^{124}I, ^{125}I, and ^{126}I contents to 0.8, 0.7, and 0.14%, respectively, by controlling the beam energy; the ^{125}I and ^{126}I are further decreased by using 99.4% isotopically enriched targets.

*Division of Medical Radiation Exposure, Cincinnati, Ohio, and †Lewis Research Center, National Aeronautics and Space Administration, Cleveland, Ohio. This article is a summary of Report BRH/DMRE 70-4, U. S. Department of Health, Education, and Welfare, Public Health Service, Bureau of Radiological Health, Rockville, Md. 20852, October 1970.

Iodine-123 was prepared by a number of nuclear reactions to determine the feasibility of different methods and the purity of the individual products. The results of these studies are reported here.

^{123}I-PRODUCTION REACTIONS

Reactions that produce ^{123}I with commonly available *n*, *p*, *d*, ^{3}He, or ^{4}He beams are shown in Table 1, and the potential targets are shown in Table 2. Because of the radioiodine contaminants in the product resulting from other isotopes in the target, enriched target material is desirable. Cost figures, however, show that only methods using ^{121}Sb, ^{123}Sb, ^{122}Te, ^{123}Te, and ^{124}Te as targets are economically feasible at present. The reactions within energy range of the small cyclotrons suitable for use in medical facilities—i.e., 7- to 10-MeV ^{2}H, 20- to 30-MeV ^{3}He, and 15- to 20-MeV ^{4}He—were especially emphasized. Both direct methods and indirect methods via ^{123}Xe, which would eliminate the longer lived high-energy radioiodine contaminants, were studied.

REACTIONS INVESTIGATED

Direct Reactions

$^{121}Sb(^{4}He,2n)^{123}I$. The E_{BP} for the $^{121}Sb(^{4}He,2n)^{123}I$ reaction is 25 MeV, which is well above the 15-MeV Coulomb barrier. The main impurity in the 98.4% ^{121}Sb target powder used was 1.6% ^{123}Sb, which yields ^{125}I from the $^{123}Sb(^{4}He,2n)^{125}I$ reaction; in addition, ^{126}I and ^{124}I are produced by the reactions $^{123}Sb(^{4}He,n)^{126}I$, $^{121}Sb(^{4}He,n)^{124}I$, and $^{123}Sb(^{4}He,3n)^{124}I$. The amount of ^{125}I produced is fixed by the ^{123}Sb impurity in the target, but the ^{124}I and ^{126}I can be minimized by using precise energy selection and extremely thin targets (10 to 15 mg/cm^2). When larger amounts of ^{123}I are desired and thicker targets (70 mg/cm^2) must be used, then, because of beam-energy attenuation, incident energies higher than the E_{BP} must be used. Furthermore, because the $(^{4}He,3n)$ reaction on ^{121}Sb produces 3.5-min ^{122}I, still higher

Table 1 Reactions for Direct Production of ^{123}I

Reaction	Coulomb barrier, MeV	E_{BP},* MeV
Direct Production		
$^{124}Xe(n,pn)^{123}I$	9	14
$^{126}Xe(p,^4He)^{123}I$	16	6
$^{120}Te(^4He,p)^{123}I$	15	16
$^{122}Te(p,\gamma)^{123}I$	8	3
$^{122}Te(d,n)^{123}I$	8	7
$^{122}Te(^3He,pn)^{123}I$	16	13
$^{122}Te(^4He,p2n)^{123}I$	15	34
$^{123}Te(p,n)^{123}I$	8	12
$^{123}Te(d,2n)^{123}I$	8	14
$^{123}Te(^3He,p2n)^{123}I$	16	20
$^{124}Te(p,2n)^{123}I$	8	21
$^{124}Te(d,3n)^{123}I$	8	25
$^{125}Te(p,3n)^{123}I$	8	28
$^{121}Sb(^3He,n)^{123}I$	15	5
$^{121}Sb(^4He,2n)^{123}I$	15	25
$^{123}Sb(^3He,3n)^{123}I$	15	20
$^{123}Sb(^4He,4n)^{123}I$	15	41
Production from ^{123}Xe (Positron Decay)		
$^{124}Xe(n,2n)^{123}Xe$	0	18
$^{120}Te(^4He,n)^{123}Xe$	15	16
$^{122}Te(^3He,2n)^{123}Xe$	16	14
$^{122}Te(^4He,3n)^{123}Xe$	15	35
$^{123}Te(^3He,3n)^{123}Xe$	16	21

*Estimated energy of the bombarding particle at which the theoretical cross section for each reaction is assumed to be maximum.

Table 2 Cost of Potential Target Materials* for ^{123}I Production (July 1, 1971)

Nuclide	Natural abundance, %	Enriched abundance, %	Cost, $/mg
^{121}Sb	57.25	95 to 99.5	2.15
^{123}Sb	42.75	95 to 99	2.50
^{120}Te	0.09	30 to 50	26.00
^{122}Te	2.46	90 to 95	2.10
^{123}Te	0.87	75 to 85	5.50
^{124}Te	4.61	>95	1.15
^{125}Te	6.99	90 to 97	1.25
^{124}Xe	0.10	10	24.35†
^{126}Xe	0.09	4	24.35†

*Antimony and tellurium enrichment and cost figures from ORNL, Isotopes Development Center, Oak Ridge, Tenn.; xenon from Mound Laboratory, Miamisburg, Ohio.

†$/ml at STP.

incident energies may be desirable to decrease the $(^4He,n)$ cross section, which produces 4.2-day ^{124}I.

The ^{123}I yields (Table 3) in the thin ^{121}Sb targets were 0.14 to 0.22 mCi/μA-hr for alpha-particle energies of 28.4 to 33.5 MeV, which is between the values 25 and 34 MeV, where the cross sections for the $^{121}Sb(^4He,2n)$ and the $^{121}Sb(^4He,3n)$ reactions, respectively, should be maximum. Energies higher than 25 MeV were used to lessen the probability of the $^{121}Sb(^4He,n)^{124}I$ reaction.

With a higher energy (36.5 MeV) bombarding particle, the ^{123}I yield was lower. The ^{124}I contamination was lowest with bombardment energies from 28.4 to 33.5 MeV, but increased with 36.5-MeV alpha particles. This increase resulted from a decrease in the probability of the $^{121}Sb(^4He,2n)^{123}I$ reaction and an increase in the probability of the $^{123}Sb(^4He,3n)^{124}I$ reaction.

With thicker ^{121}Sb targets—50 mg/0.7 cm^2—the ^{123}I yields were 0.38 to 0.93 mCi/μA-hr with an alpha-particle bombardment energy of 31 ± 3 MeV, but the average yield (0.012 mCi/μA-hr per milligram) was the same as for 15 mg/0.7 cm^2 targets. The ^{124}I contamination was 0.65 to 1.06%, averaging 0.79 ± 0.13%, which is 30% higher than that with the 15 mg/0.7 cm^2 targets. The higher ^{124}I contamination resulted from the increased probability of the $^{121}Sb(^4He,n)^{124}I$ reaction occurring as the alpha-particle energy was attenuated in traversing the thicker targets. However, the approximately linear relation of ^{123}I yield vs. the thickness of the ^{121}Sb targets indicated that, in penetrating the 50 mg/0.7 cm^2 target, the incident alpha-particle energy (34 MeV) was not attenuated below that which caused the desired $^{121}Sb(^4He,2n)^{123}I$ reaction. Probably the ^{123}I yield per μA-hr could have been increased by using even thicker ^{121}Sb targets but, unfortunately, the ^{124}I contamination would also have been increased.

Table 3 Radioiodine Yields from the $^{121}Sb(^{4}He,2n)^{123}I$ Reaction on Thin (~15 mg/0.7 cm^2) Targets of ^{121}Sb

Energy, MeV	Target thickness, mg/0.7 cm^2	^{123}I, mCi/μA-hr	Contaminant, %		
			^{124}I	^{125}I	^{126}I
28.4 ± 1.2	15.0	0.14	0.54	0.014	0.0023
33.3 ± 1.1	15.8	0.23	0.62	0.015	0.0019
33.4 ± 1.0	15.0	0.18	0.53	0.013	0.0029
33.5 ± 0.9	13.4	0.16	0.54	0.014	0.0023
36.5 ± 1.0	15.0	0.08	1.35	0.014	0.0022

$^{121}Sb(^{3}He,n)^{123}I$. The Coulomb barrier for this reaction, 15 MeV, is 10 MeV higher than its E_{BP}. However, although this reaction has a low theoretical cross section, it was attempted because extremely high-purity ^{121}Sb is available. Further, the 15-MeV ^{3}He particles can be obtained from the low-cost cyclotrons available to many nuclear medical institutions, and, theoretically, no ^{124}I should be produced from the $(^{3}He,\gamma)$ reactions, which have low probability with incident 5- to 15-MeV ^{3}He particles.

However, the results showed that this reaction cannot be used for production of radiopharmaceutical-grade ^{123}I. With thin ^{121}Sb targets and 4.5-MeV ^{3}He particles, the ^{123}I yield was only 8.5×10^{-6} mCi/μA-hr, due to limitation by the Coulomb barrier. The ^{124}I contamination was 2.7%. With 14-MeV bombarding particles the ^{123}I was 10 times as high, but the ^{124}I increased to 4.3%, and a large amount of ^{121}I was formed from the $^{121}Sb(^{3}He,3n)^{121}I$ reaction.

At this energy the cross sections for the $(^{3}He,2n)$ and $(^{3}He,3n)$ reactions have increased more than that of the desired $(^{3}He,n)$ reaction. Undoubtedly, ^{122}I was also produced by the $^{121}Sb(^{3}He,2n)$ reaction but had decayed by the time of analysis.

$^{122}Te(d,n)^{123}I$. The Coulomb barrier for the $^{122}Te(d,n)^{123}I$ reaction, 8 MeV, is close to the E_{BP}, 7 MeV, and the reaction was attempted at 6.4 to 9.0 MeV. In favor of this reaction is the limitation of the radioiodine contamination to (d,n) and $(d,2n)$ reactions on tellurium impurities in the target, with no competing reactions on ^{122}Te. The $^{122}Te(d,\gamma)^{124}I$ reaction is highly improbable at these bombardment energies. The $^{122}Te(d,2n)^{122}I$ reaction produces the 3.5-min contaminant. Inquiries to two suppliers of ^{122}Te showed that it was economically impractical to reduce the tellurium impurities to less than 5%.

The ^{123}I yields (Table 4) from 6.8- to 7.3-MeV deuteron bombardment of 15.8 to 31.5 mg/0.7 cm^2 ^{122}Te targets varied from 0.015 to 0.132 mCi/μA-hr. This large variation is probably related to the fact that the calculated Coulomb barrier for the reaction is approximately 8 MeV. Assuming that the cross section of the $^{122}Te(d,n)^{123}I$ reaction is a smooth function of the excitation energy, it was estimated that the cross section should be approximately maximum at the theoretical threshold for the $^{122}Te(d,2n)^{122}I$ reaction, i.e., about 7 MeV. However, because the Coulomb barrier opposes the approach of the deuteron to the ^{122}Te nucleus at this energy, the cross section for the reaction will probably actually be a very steep function of the energy, as borne out by the 0.0019 mCi/μA-hr ^{123}I yield obtained at 6.4 MeV. The large variations in ^{123}I yields under seemingly identical conditions are probably due to the unavoidable variations in the thicknesses of the powder targets, which in some instances decrease the excitation energy to values significantly lower than the Coulomb barrier.

The ^{123}I yield with 9.0-MeV deuteron-bombardment energy, about 1 MeV higher than the calculated Coulomb barrier, was approximately the same as that obtained at 6.8 to 7.3 MeV. The yields of ^{124}I, ^{126}I, and ^{130}I, however, were twofold to threefold higher at this higher excitation energy because of the increased cross sections of the $^{124}Te(d,2n)^{124}I$, $^{126}Te(d,2n)^{126}I$, and $^{130}Te(d,2n)^{130}I$ reactions. The results show that the highest and purest yields of ^{123}I were obtained with 7-MeV deuteron bombardment of 15 mg/0.7 cm^2 targets of 95.4% ^{122}Te.

Indirect Reactions

$^{122}Te(^{4}He,3n)^{123}Xe + {}^{122}Te(^{4}He,p2n)^{123}I$. The E_{BP} for these reactions, 35 MeV, is well above their Coulomb barriers (15 MeV). These reactions were studied together by entrapping the ^{123}Xe gas in the sealed target along with the directly produced ^{123}I.

Table 4 Radioiodine Yields from the $^{122}Te(d,n)^{123}I$ Reaction on 95.4% ^{122}Te

Energy, MeV	Target thickness, mg/0.7 cm²	^{123}I, mCi/μA-hr	Contaminant, %				
			^{124}I	^{125}I	^{126}I	^{130}I	^{131}I
6.4 ± 0.3	12.5	0.0019	0.072	0.016	0.041	0.97	0.15
6.8 ± 0.5	17.3	0.015	0.083	0.021	0.039	0.92	0.07
6.8 ± 0.5	16.3	0.101	0.085	0.013	0.044	0.86	0.09
6.9 ± 0.4	15.8	0.062	0.072	0.021	0.034	0.82	0.08
7.3 ± 1.0*	31.5	0.132	0.192	0.012	0.066	0.96	0.07
9.0 ± 0.3	12.6	0.081	0.240	0.013	0.096	2.5	0.09

*Bombardment performed on 80.4% ^{122}Te and yields corrected to 95.4% ^{122}Te.

The sealed target was opened after the ^{123}Xe had decayed to ^{123}I.

The ^{123}I yield from 40-MeV ^{4}He reactions on ^{122}Te was about 0.012 mCi/μA-hr per milligram of target, 17 to 36 mg/cm² thick (Table 5). The incident alpha particles at 41 MeV will decrease in energy to 34 MeV in penetrating a 75 mg/cm² ^{122}Te target, and if 35 MeV is the E_{BP} for these reactions, we can assume a linear increase of ^{123}I yield with targets at least up to 75 mg/cm² in thickness. Therefore the projected ^{123}I yield from these reactions is about 0.65 mCi/μA-hr.

$^{122}Te(^{3}He,2n)^{123}Xe + ^{122}Te(^{3}He,pn)^{123}I$. The Coulomb barriers for $^{122}Te(^{3}He,2n)^{123}Xe$ and $^{122}Te(^{3}He,pn)^{123}I$ reactions at 16 MeV are higher than the E_{BP}'s, 14 and 13 MeV, respectively, and these reactions would be expected to have limited yields. However, the $^{122}Te(^{3}He,2n)^{123}Xe$ reaction was attempted because it was within the energy limitations of the compact cyclotron. The compact cyclotron cannot accelerate alpha particles to the 35 to 40 MeV necessary to cause the $^{122}Te(^{4}He,3n)^{123}Xe$ reaction.

These reactions were studied (1) by allowing the bombarded target to decay several hours and then chemically separating the ^{123}I, yields from both reactions being corrected to the time of maximum ^{123}I ingrowth from ^{123}Xe, or (2) by counting bombarded targets without chemical treatment to individually assay the $^{122}Te(^{3}He,2n)^{123}Xe$ and $^{122}Te(^{3}He,pn)^{123}I$ reactions and then calculating the total ^{123}I yield at the optimum ingrowth time from the individual yields.

The ^{123}I yields (Table 6) from 9.8 and 13.8 mg/0.7 cm² sealed ^{122}Te (80.4%) targets were 0.023 and 0.018 mCi/μA-hr, respectively. The ^{124}I contamination averaged 0.7%. With 73.3% targets, the maximum amount of ^{123}I (Table 6) at optimum ingrowth time was a factor of 100 less at 13.5 MeV than that at 17.9 MeV due to factors related to the 16-MeV Coulomb barrier for this reaction.

The ^{123}Xe and ^{123}I yields per milligram of target at 17.9 MeV were, respectively, 100 and 150 times that at 13.5 MeV. This can be explained by the increasing probability of charged-particle emission from the compound nucleus at the higher energy. To compare the ^{123}I yields from the direct and indirect sources, the ^{123}Xe yields must be divided by 6.3, i.e., $T_{1/2}(^{123}I)/T_{1/2}(^{123}Xe)$. The direct ^{123}I yield from the $^{122}Te(^{3}He,pn)^{123}I$ reaction is about two to three times the indirect yield from the $^{122}Te(^{3}He,2n)^{123}Xe$ reaction.

$^{123}Te(^{3}He,3n)^{123}Xe + ^{123}Te(^{3}He,p2n)^{123}I$. The E_{BP}'s of the $^{123}Te(^{3}He,3n)^{123}Xe$ and $^{123}Te(^{3}He,p2n)^{123}I$ reactions at 21 and 20 MeV, respectively, are well above their 16-MeV Coulomb barriers.

In favor of this reaction over other ^{123}Xe producing reactions—i.e., $^{122}Te(^{4}He,3n)^{123}Xe$ and $^{122}Te(^{3}He,2n)^{123}Xe$—are the following:

1. The 25-MeV helium particle necessary for this reaction is within the energy limitations of some compact cyclotrons, whereas the 42-MeV ^{4}He particle necessary for the $^{122}Te(^{4}He,3n)^{123}Xe$ reaction is not.
2. Because the ^{3}He reaction with ^{123}Te and the ^{4}He reaction with ^{122}Te form the same compound nucleus, ^{126}Xe, yields should be identical.
3. No Coulomb-barrier interference exists as there does with the $^{122}Te(^{3}He,2n)^{123}Xe$ and the $^{122}Te(^{3}He,pn)^{123}I$ reactions.
4. Because the desired ^{3}He-induced reactions on ^{123}Te require higher energy ^{3}He particles than do those on ^{122}Te, thicker targets can be used.

Table 5 Radioiodine Yields from the $^{122}Te(^{4}He,3n)^{123}Xe \rightarrow {}^{123}I$ and $^{122}Te(^{4}He,p2n)^{123}I$ Reactions on 95.4% ^{122}Te

Energy, MeV	Target thickness, mg/0.7 cm²	^{123}I, mCi/μA-hr	Contaminant, %		
			^{124}I	^{125}I	^{126}I
40.4 ± 0.7	11.3	0.16	1.1	0.03	0.006
40.0 ± 1.1	17.2	0.21	1.6	0.06	0.004
39.5 ± 1.6	25.0	0.25	1.5	0.03	0.009

Table 6 Yields from the $^{122}Te(^{3}He,2n)^{123}Xe$ and $^{122}Te(^{3}He,pn)$ Reactions

Energy, MeV	Target wt., mg/0.7 cm²	Yield			
		^{123}Xe, mCi/μA-hr	^{124}I, %	^{123}I, μCi/μA-hr	Total ^{123}I, μCi/μA-hr
80.4% ^{122}Te Target					
14.7 ± 0.9	9.8		0.63	0.023	
14.2 ± 1.4	13.8		0.83	0.018	
73.3% ^{122}Te Target					
13.5 ± 0.8	7.8	0.18		0.06	0.07
17.9 ± 0.6	5.4	13.7		6.3	6.7

The disadvantages of reactions using ^{123}Te compared to those using ^{122}Te are the higher cost and the lower isotopic enrichments of ^{123}Te.

The maximum available ^{123}I yields calculated to the time of optimum ingrowth (Table 7) from the ^{3}He-induced reactions on ^{123}Te were only 2% of that from the ^{4}He-induced reactions on ^{122}Te (Table 5). However, the ^{123}I yields from ^{3}He-induced reactions on ^{123}Te could be increased by using particles higher in energy than the 22-MeV ^{3}He available for this work. These reactions will be further studied at 28 MeV. Assuming that the compound nucleus ^{126}Xe is formed with both the ^{3}He-induced reactions on ^{123}Te and the ^{4}He-induced reactions on ^{122}Te, comparable ^{123}Xe and ^{123}I yields would be expected.

Table 7 ^{123}Xe, ^{123}I, and Total Available ^{123}I from the $^{123}Te(^{3}He,3n)^{123}Xe$ and $^{123}Te(^{3}He,p2n)^{123}I$ Reactions on 76.5% ^{123}Te

Energy, MeV	Target thickness, mg/0.7 cm²	Yield, μCi/μA-hr		
		^{123}Xe	^{123}I	Total ^{123}I
16.8 ± 0.5	5.4	1.6	0.56	0.60
21.4 ± 0.4	6.0	7.7	0.78	1.44

An interesting observation is that the ^{123}Xe yield increased fivefold at the higher energy whereas the ^{123}I yield increased only 39%. If this trend continues at still higher energies, most of the available ^{123}I would be produced indirectly from ^{123}Xe decay.

Gas-Flow Target Assembly

The $^{122}Te(^{4}He,3n)^{123}Xe$ reaction was tested with a continuous gas-flow target. Isolation of xenon from ^{3}He and ^{4}He bombardment of tellurium is feasible for pure ^{123}I production because:

1. Xenon can be physically separated from other elements.
2. The ^{124}Xe, ^{126}Xe, ^{128}Xe, ^{129}Xe, ^{130}Xe, ^{131}Xe, ^{132}Xe, and ^{134}Xe isotopes are stable.
3. The 36.4-day ^{127}Xe and 5.3-day ^{133}Xe isotopes decay to stable daughters.
4. Xenon isotopes with atomic mass less than 122 decay with short half-lives (≤41 min) to short-half-life radioiodine daughters (≤2 hr).
5. The 19-hr ^{122}Xe decays to 3.5-min ^{122}I.
6. The major radioiodine contaminant is 60-day ^{125}I, which results from decay of 17-hr ^{125}Xe. Separation of ^{123}I from ^{123}Xe after only 2- to 5-hr decay prevents the ingrowth of most of the available ^{125}I.

Furthermore, the gas-flow method is advantageous over the conventional sealed target–chemical separation method for these additional reasons:

1. The ^{123}Xe decays to monatomic ^{123}I, which is chemically very reactive and presents opportunities for the direct labeling of organ-specific compounds.
2. The time necessary for ^{123}I ingrowth can be used to transport the isotope to the nuclear medical facility.
3. The target material does not have to be recovered.
4. The preparation of radiopharmaceutical ^{123}I by simply leaching the trap eliminates the standard iodine distillation.
5. The handling of non-^{123}I radioactivity normally encountered with bombardment of standard targets is not necessary.

The ^{123}I yield from 100 to 127 mg of 95.4% ^{122}Te spread over an area of 6 cm^2 averaged about 0.045 mCi/μA-hr. The yield from 10 g of natural tellurium in the gas-flow target was 0.003 mCi/μA-hr.

In one run with a helium flow rate of 10 liters/min and a copper U trap to remove contaminants, the ^{124}I was 0.27%; however, with a 10-turn copper coil the ^{124}I was only 0.05%. Better target assemblies using more efficient water cooling of the powder could probably be devised.

Allowing the xenon trap to decay only 3 to 4 hr optimized the ^{123}I yield and kept ^{125}I ingrowth to <0.2%.

CONCLUSIONS

Direct Methods

The ^{121}Sb(^{4}He,2n)^{123}I reaction will provide satisfactory yields, 0.60 mCi/μA-hr, for most needs. However, the 0.8% ^{124}I contamination limits the product's usefulness in high-resolution nuclear medical applications, and the need for 34-MeV ^{4}He particles restricts the production to higher energy cyclotrons.

The ^{122}Te(d,n)^{123}I reaction with 7-MeV deuterons on 95.4% ^{122}Te gives high-energy long-lived radioiodine contaminants of only 0.3% and of ^{130}I, 0.9%. This method is useful with a compact cyclotron, but the yield is limited to about 0.1 mCi/μA-hr. The short range of 7-MeV deuterons in tellurium restricts target thickness to less than 25 mg/cm^2.

The ^{121}Sb(^{3}He,n)^{123}I reaction is not practical at any energy. The ^{122}Te(^{4}He,$p2n$)^{123}I, ^{122}Te(^{3}He,pn)^{123}I, and ^{123}Te(^{3}He,$p2n$)^{123}I reactions are of doubtful usefulness since the ^{124}I contamination of all these products is >1%.

Indirect Methods

The ^{122}Te(^{4}He,3n)^{123}Xe reaction produces the highest yield of pure ^{123}I, 0.2 mCi/μA-hr, with 41-MeV ^{4}He particles on 75 mg/cm^2 95.4% ^{122}Te. However, no compact cyclotron will accelerate 40-MeV ^{4}He particles.

The ^{122}Te(^{3}He,2n)^{123}Xe reaction requires <20-MeV ^{3}He particles, which can be produced with the compact cyclotron. However, because of the Coulomb-barrier and range-energy considerations, the yield is too low for practical use.

The ^{123}Te(^{3}He,3n)^{123}Xe reaction has no Coulomb-barrier problem and needs a 28-MeV ^{3}He beam, which is available from some compact cyclotrons. The yield should be comparable to the ^{122}Te(^{4}He,3n)^{123}I since the compound nucleus, ^{126}Xe, is the same. (Since only a 22-MeV beam was available, this was not verified.) (MG)

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Instant ^{99m}Tc Compounds

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Supplementary Keywords: *medicine; diagnosis; industry, pharmaceutical; preparation, labeled material;* ^{99m}*Tc.*

Abstract: *For the rapid production of high-purity* ^{99m}*Tc radiopharmaceuticals, stannous ion was used as a reducing agent for technetium. Procedures for two such "instant" compounds are reported.*

One goal of the radiopharmaceutical-development program at Brookhaven National Laboratory (BNL) is to provide ^{99m}Tc compounds which contain a single component and which can be easily prepared, ideally by adding pertechnetate to a prepared solution. The one-step preparation of single-component products would lead to the widespread use of these compounds, especially if they were commercially available to hospitals and clinics that have limited radiopharmaceutical facilities. Further, sterility and pyrogenicity could be determined before use.

Clinical evaluation[1,2] has indicated that the chelate ^{99m}Tc–Fe–DTPA (diethylenetriaminepentaacetic acid) is superior to ^{99m}Tc pertechnetate, the ^{99}Mo generator product, for brain scanning and kidney studies, and this compound has been prepared at BNL. However, considerable time and manipulation are necessary for the preparation, and a one-step preparation for high-purity products should greatly increase the use of technetium and help make the most advanced diagnostic techniques available to a large cross section of hospitals and clinics. Stannous ion has been found an effective reducing agent in the labeling of red blood cells.[3] It was used for reducing the $^{99m}TcO_4^-$ to a cationic ^{99m}Tc species that is incorporated in compounds,[4] and rapid and simple procedures were developed for making ^{99m}Tc–DTPA and ^{99m}Tc–HSA (human serum albumin). These are described here. Development of kits, using similar procedures, for other biologically interesting compounds may be possible.

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PREPARATION OF ^{99m}Tc–DTPA

The method for making "instant" ^{99m}Tc–DTPA is based on the preparation of a stannous DTPA chelate that reduces and binds technetium at neutral *p*H. A DTPA/$SnCl_2$ mole ratio of about 8/1 should be sufficient to yield a stable complex.[5] With this ratio, the amount of tin chloride administered per patient would be about 125 μg, which is not toxic.[6]

The nonradioactive chelate is prepared first, as a stock solution, in amount sufficient for about 40 patients. The stock solution, which is stable for at least 14 days at 4°C, is made under sterile conditions and using pyrogen-free reagents, as follows:

1. To 1 ml of $CaNa_3$–DTPA solution, add 5 mg of $SnCl_2 \cdot 2H_2O$ and heat at 100°C under N_2 for 15 min.
2. Dilute to 18 ml with H_2O, adjust *p*H to 4.0 with HCl, and dilute to 20 ml with H_2O.
3. Purge solution with N_2 for 15 min and then filter through a 0.22-μ filter into vials under N_2, 1 ml of solution per vial.

A DTPA concentration of at least 100 mg/ml in the starting solution is necessary for good dissolution of $SnCl_2 \cdot 2H_2O$ at *p*H 4. The stock solution can be diluted without precipitation of the tin.

For administration to a patient, up to 3 ml of saline pertechnetate solution is added to 1 ml of stock solution and mixed for 1 min. This gives the injectable amount desired in a reasonable volume. No purification is necessary because the chelate is formed in greater than 95% yield.

PREPARATION OF ^{99m}Tc–HSA

In developing a simple procedure for ^{99m}Tc–HSA, the approach was similar to that used for ^{99m}Tc–DTPA. Stannous HSA chelate was prepared at various *p*H values, but, unlike the DTPA case, the bulk of the tin could not be chelated with up to 250 mg of HSA (about 3 μmoles). With 60 μg of $SnCl_2 \cdot 2H_2O$, only 23% appeared bound to 250 mg of HSA at *p*H 6, the rest being present as colloid. To prevent competition for the ^{99m}Tc between the colloid and the HSA, the ^{99m}Tc is added to the $SnCl_2$–HSA solution at *p*H 2. The stock solution, which is stable for 10 days

when stored under N_2 at 4°C, is prepared, using similar precautions against contamination as in the preceding preparation, as follows:

1. Dissolve 12.5 mg of $SnCl_2 \cdot 2H_2O$ in 1 ml of concentrated HCl with heat and dilute to 25 ml with N_2-purged H_2O.
2. Store under N_2 in 10-ml capped vials, 0.5 ml (250 μg of $SnCl_2 \cdot 2H_2O$) per vial.

A ^{99m}Tc–HSA solution is made in ~95% yield, with high reproducibility, by adding 1 ml of HSA solution (250 mg/ml) to 0.5 ml of the stock tin solution and then 0.1 to 2 ml of $^{99m}TcO_4^-$ solution, mixing for 1 min, and finally adding 0.6 ml of 0.8*M* Na_2HPO_4. This is not actually a one-step procedure because the tin–HSA combination cannot be stored, and the *p*H of the final solution must be adjusted. Nevertheless, it is a relatively simple technique and is more satisfactory than one-step preparations made by using less stannous ion initially or by gel-chromatographic removal of the tin colloid before addition of the pertechnetate, both of which techniques were developed. (MG)

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SUMMARY OF FEDERAL REGULATIONS FOR PACKAGING AND TRANSPORTATION OF RADIOACTIVE MATERIALS

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Various governmental regulations on packaging and transportation of radioactive materials are summarized. Included are definitions of terms; classifications of radioactive materials; general and specific requirements, quantity limitations, and labels for radioactive material packages; and information on shippers' and carriers' responsibilities and on loading and storage of radioactive materials. (MG)

Section II

Isotopes and Radiation Technology

Isotope Technology Development

Radioanalytical Methods Applied to Air–Sea Interaction Studies

By P. E. Wilkniss, D. J. Bressan, and R. E. Larson*

Supplementary Keywords: *activation, neutron; activation, charged particle; activation, photon; production, radioisotope; production, reactor; production, accelerator; chemistry, analytical; meteorology; oceanology; hydrology; material tracing; spectrometry; pollution; atmosphere; data handling; differentiation;* ^{18}F; ^{24}Na; ^{38}Cl; ^{42}K.

Abstract: *Evidence is presented that activation analysis is an excellent way to determine the composition of aerosols. Elemental ratios derived therefrom can distinguish between oceans and continents as the sources of aerosols and have potential for differentiating airborne continental dust, volcanic dust, and polluted air.*

A laboratory experiment with radionuclide tracers simulated the bubble-bursting process whereby particulates are transferred from the ocean to the atmosphere. The results, compared with those for real marine aerosols, show that the technique should be useful for laboratory-scale study of the environment.

Large-scale interactions between the oceans and the atmosphere exert a driving force on atmospheric circulation and influence global weather and climates. Obviously, a thorough understanding of the ocean–atmosphere boundary layer and of the processes that occur in it is very important in long-range weather forecasting, planning weather modifications (e.g., seeding hurricanes), and understanding climatic changes caused by human activities, such as pollution of the atmosphere or of the ocean surface.

Scientific investigation of the air–sea interface is a formidable problem that requires interdisciplinary effort by marine biologists, chemical and physical oceanographers, meteorologists, cloud physicists, and atmospheric chemists. "Environmental chemists" have contributed significantly in this area. Chemical methods will be needed in the future to establish whether the oceans are sinks or sources for certain atmospheric constituents, to study atmospheric circulation over the oceans, to investigate interhemispheric transport, to learn more about chemical reactions in the marine atmosphere and in oceanic clouds, to investigate the existence of a "background aerosol" over the ocean, and to trace the migration of man-made pollutants into the marine atmosphere and the oceans. Radiochemistry has played an important role in these investigations, mainly through the natural radioactivity and bomb-produced fission products that occur in the oceans and the atmosphere.

Activation analysis and man-made radionuclides have been used increasingly in environmental studies. A summary is given below of work to determine the chemical composition and origin of marine-atmospheric particulates and to simulate in the laboratory, by use of radionuclides, the transport of inorganic particulates from the oceans into the atmosphere.

ACTIVATION ANALYSIS

The ideal method for analyzing marine-environmental samples should be (1) flexible to sample type (seawater, sea spray, rain, ice, snow, and continental or volcanic dust); (2) capable of multielement determinations to permit measuring ratios of several elements; (3) highly sensitive (microgram samples that contain less than nanograms of trace elements); (4) without need for chemical pretreatment, thus avoiding the introduction of contaminants; and

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(5) nondestructive so as to preserve unique samples for further examination. Activation analysis fulfills all these requirements. Even the economic aspects of activation analysis, which often make it noncompetitive, are less restrictive in this case.

Unique samples and those from remote locations are precious. In general, sampling that requires the use of aircraft and ships is very expensive and thus partly justifies the use of more costly methods of analysis. Because of the complex analytical problem that environmental samples present, no one type of activation analysis can satisfy all the requirements. Therefore, examples of the use of activation analysis with neutrons, photons, and protons are presented.

With Neutrons

Nondestructive neutron activation analysis was used to determine Al, Ce, Co, Eu, Fe, Mg, Mn, Na, Sb, Sc, Ti, and V in airborne dust collected over the Atlantic Ocean.[1] After short and long irradiation, the samples were counted by use of a Ge(Li) detector and a 4096-channel analyzer. Computer-aided evaluation and comparisons with standard samples gave elemental ratios for environmental samples from several sources (pumice M-1 from the 1965 eruption of Kilauea Volcano, Hawaii; seawater; and polluted urban air from U. S. cities).[2,3] Figure 1 includes Mg/Fe, Ti/Fe, V/Fe, and Na/K ratios.

With Photons

Photon activation analysis is an excellent method to determine fluorine and chlorine in seawater, marine aerosols, and airborne dust.[4] To meet the stringent geometry requirements for uniform photon irradiation, liquid samples must be freeze-dried. Irradiation at a maximum photon energy of 22 MeV avoids the interfering reactions $^{23}Na(\gamma,\alpha n)^{18}F$ and $^{18}O(p,n)^{18}F$. Good sensitivities for fluorine and chlorine are obtained by counting with a low-background coincidence arrangement. Figure 1 gives results from Ref. 5 for the simultaneous determinations, by photon activation, of fluorine and chlorine in dust from the vicinity of Mid-Atlantic Ridge, marine aerosols from the Atlantic and Pacific Oceans, and coastal aerosols collected ~30 miles off the coast of New Jersey. These results represent only a few analyses on very good samples (low filter blanks).

With Protons

A preliminary study[6] showed that the activation of sea salt with 10-MeV protons permits the simultaneous determination of Br, Ca, S, and Sr, without radiochemical separation, by use of a Ge(Li) detector and a 4096-channel analyzer. The (p,n) reactions at 10 MeV do not produce interfering nuclear-reaction products of Na, Cl, and Mg, which are the main constituents of sea salt. To date, no atmospheric samples have been analyzed by this method; therefore, no results are presented.

Discussion

Activation analysis of environmental samples is not yet very precise. In unfavorable cases, relative standard deviations may be ±50% or higher, mainly because of high blanks from the filters used to collect samples. To positively identify samples by elemental ratios, the ratios should differ by at least an order of magnitude. The Na/K and F/Cl ratios for marine aerosols are very close to those for seawater (Fig. 1), an agreement which strongly suggests that the ocean is the source of these aerosols. The F/Cl, Mg/Fe, and V/Fe ratios for the Atlantic dust samples clearly identify the continents (crustal rock)—not the oceans—as the source of this material. The Ti/Fe ratio for the Atlantic dust indicates that none of the dust is of volcanic origin, but more evidence is needed. Continental dust and polluted urban air are much more difficult to differentiate as the source of particulates in a "coastal" aerosol—i.e., an aerosol collected over the ocean—but near (30 to 50 miles) an industrialized area on land. Again, the F/Cl ratio seems to be useful, but many more ratios, especially metal ratios (e.g., V/Fe), are definitely needed. Also, the determination of sulfates or other sulfur-containing species that predominate in polluted urban air would be useful.

The conclusion is that activation analysis can easily identify the source of an aerosol as being either the oceans or the continents. The method shows good potential for differentiating among airborne continental dust, volcanic dust, and polluted air, especially when a successful procedure for sulfur compounds is included.

TRANSPORT OF PARTICULATES FROM THE OCEAN INTO THE ATMOSPHERE—A LABORATORY INVESTIGATION WITH RADIOTRACERS

The quantity of inorganic particulates transported from the world oceans into the atmosphere is estimated to be 10^9 metric tons/year. This tremendous amount of salt is injected into the air mostly in the form of tiny sea-spray droplets, which are produced by

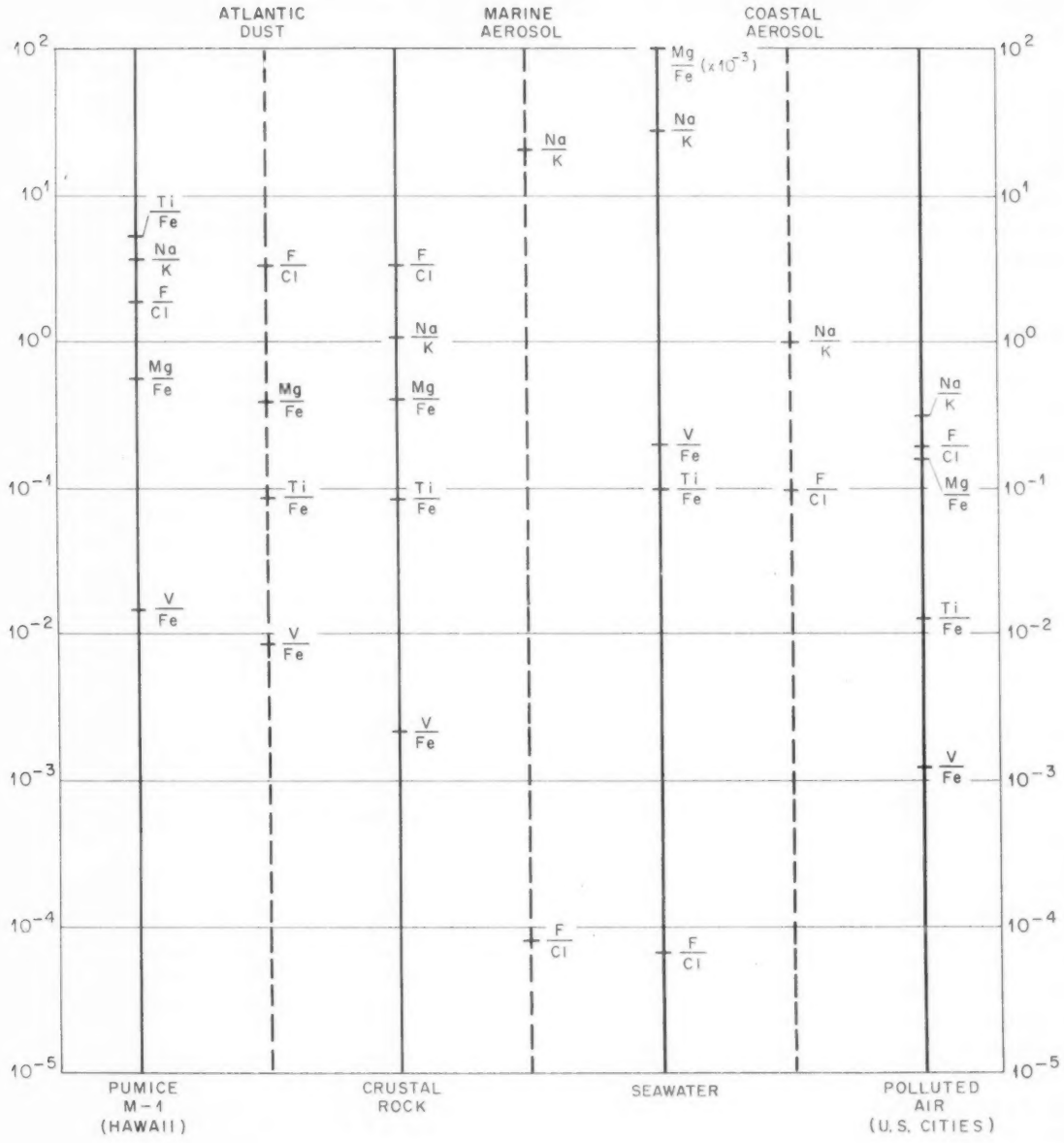

Fig. 1 Elemental ratios for environmental samples. - - -, based on activation-analysis data from this study. ——, based on data from the literature.

the bursting of small bubbles at the ocean surface—for instance, in the whitecaps of wind-driven waves. The droplets dry out quickly, stay airborne as salt particles, and are carried away by the wind.[7]

Obviously, this bubble-bursting process cannot be simulated easily in the laboratory simply by bubbling air through a beaker that contains seawater. However, with a sensitive means to detect the very small amounts of sea-salt particles produced, "microoceans" can be studied in the laboratory. Radiotracers are extremely well suited to this application. In this approach, several elements in the seawater being studied are labeled with radiotracers, air is bubbled through the solution, and the seawater and sea spray are analyzed by gamma-ray spectroscopy. The method has the following advantages: (1) use of sufficient amounts of radionuclides yields measurable samples after bubbling periods that approach those of real oceanic processes; (2) use of carrier-free tracers permits work with trace elements, at very low concentrations, that cannot be determined by any other analytical method; (3) the behaviors of elements present in different chemical forms or valence states can be studied following their injection into the atmosphere; (4) chemical processes that occur at the air–sea interface and within the salt particles in the atmosphere can be studied; and (5) the experiments are nondestructive and can be repeated under different conditions.

A practical example illustrates the experimental possibilities. A 50-ml sample of seawater was labeled with ^{18}F, ^{24}Na, ^{38}Cl, and ^{42}K. The ^{18}F was produced carrier free in a cyclotron; the other three radionuclides were formed in high specific activity by neutron irradiation of *Na*Cl, NH_4Cl, and *K*Cl in a reactor. All four radionuclides were introduced into the seawater by a straightforward chemical operation.[7]

The bubbling experiment was conducted in a humid enclosure to maintain the relative humidity about that of the real ocean surface. The labeled seawater was contained in a fritted-glass cylinder inside the enclosure. Air was bubbled through the frit. The bursting bubbles produced fairly large sea-spray drops ("jet drops"), which were collected on an aluminum-foil collar around the glass cylinder. Much-finer particles were also produced; they filled the enclosure with a radioactive sea-salt aerosol. The aerosol was sampled by vacuum filtration of the air in the enclosure through a 0.45-μ Millipore filter. Gamma-ray spectrometry was used to measure the radioactivity in the original seawater, in the jet drops, and in the aerosol. Table 1 gives the results, expressed as elemental ratios.

Table 1 Elemental Ratios Measured in Bubble-Bursting Experiments with Labeled Seawater*

Sample	Elemental ratio					
	F/Cl	F/Na	F/K	Cl/Na	Cl/K	Na/K
	$\times 10^{-5}$	$\times 10^{-4}$	$\times 10^{-3}$		$\times 10$	$\times 10$
Labeled seawater	6.7	1.2	3.4	1.8	5.0	2.8
Jet drops	6.4	1.2	3.2	2.0	5.5	2.2
Aerosol	1.1	0.3	-	2.0	-	-
Marine aerosols†	9.0	0.9	-	0.8	-	1.8

*The ratios given are not activity ratios but rather calculated elemental ratios, using those in seawater of 35‰ salinity as basis.

†Values for marine aerosols (see Fig. 1) are given for comparison.

Discussion

Except for the Cl/Na value, all the elemental ratios for laboratory-produced jet drops agree satisfactorily with those for the aerosols collected over the ocean. The agreement indicates that jet drops produced in the laboratory are similar to those which form over the real oceans. However, for the laboratory aerosol, both the F/Cl and F/Na ratios were much lower than those for the natural aerosol. This result indicates a chemical fractionation among F, Cl, and Na during the processes that occur at the air–sea interface.[5] The fractionation seems to occur only for the very small particles of the aerosol. So far, not enough of these small particles have been collected from the marine atmosphere to verify the laboratory results. This effect was not observed for the regular aerosols, which are dominated by the mass of large particles (sea-spray drops).

These findings have twofold importance for oceanographic studies: they show that (1) with this method, it is possible to observe and to study in the laboratory those processes such as chemical fractionation which occur at the air–sea interface; and (2) radioactively labeled aerosols that simulate those aerosols which occur in nature over the oceans can be produced and investigated. The method is not limited to oceanographic applications but is generally applicable. Multiply labeled aerosols can be produced from almost any solution of radiotracers by air bubbling—for instance, labeled aerosols from sulfuric acid solution to resemble pollutant aerosols found in urban atmospheres. The method may be useful in air chemistry as well as in medical research, such as studies of the elimination of

particles from human lungs. Radionuclides can be used in aerobiological studies—e.g., study of the production and distribution of infectious aerosols from bacteria-containing solutions or the spread of such aerosols from aerated settling lagoons. Since the falling of drops on shallow liquids produces similar aerosols, the spread of bacteria-containing aerosols from trickling-filter sewage plants can be investigated.[8] Also, the transport of pollutants into the atmosphere from any contaminated water body in which bubbles are formed can be followed. Examples of such studies are the injection, into the atmosphere, of oil pollution from films on the sea surface and of raw sewage (dissolved or particulate form) from a sewage-discharge location.

CONCLUSIONS

Activation analysis is an excellent method to determine the chemical composition of aerosol particles from the marine atmosphere. However, further work is needed to eliminate the problem of high blanks posed by present sampling techniques. Application of the method at its greatest sensitivity is mandatory to obtain and improve determinations of the chemical composition of particles of different sizes, especially up to 1 μ. A good activation-analysis technique is needed to determine sulfur because of the great importance of this element in pollution and environmental studies.

The bubble-bursting process—the main source of salt particles over the oceans—in combination with radiotracers offers many possibilities for laboratory study of the environment. To label natural waters without disturbing the existing delicate chemical equilibria is difficult. However, many elements can be labeled carrier free without such problems. In addition to oceanographic investigations, activation analysis has great potential for aerobiological and pollution studies. It can be performed with simple equipment in any laboratory and can be adapted easily to small-scale field work.[7] (HPR)

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DETECTION OF MATERIALS REMOVED FROM AN INDUSTRIAL PLANT USING RADIOACTIVE TRACERS

By A. C. Castagnet, H. Rodolfo Gomez, and A. H. Lammirato,
Argentinian Report CNEA–EN–1/6 (in Spanish), September 1968.

Supplementary Keywords: *forensic use; material tracing; industry;* ^{110m}Ag.

At the request of a local factory manager, a method was developed for ^{110m}Ag labeling of copper piping and mercury so that these materials could be detected if removed from the plant without authorization. The mercury was labeled (0.5 μCi/g) by adding the ^{110m}Ag as an amalgam, and the copper piping (5 μCi/cm), by depositing $^{110m}AgNO_3$ on a surface previously treated to provide redox conditions. The detection system—Geiger–Mueller counters connected to an alarm system—was placed in the plant exit corridors and could detect 300 g of mercury and 10 cm of copper piping. The cost for installing a detection system in two corridors was estimated at $4000. The levels of activity involved were not considered to be a health hazard. (MG)

Automatic Industrial Activation-Analysis System for Slurries Using a Radioisotope Neutron Source*

By Kenneth R. Blake, John B. Ashe, Peter F. Berry, and John B. Nelson†

Supplementary Keywords: *activation analysis; activation, neutron; chemistry, analytical; metallurgy, ferrous; industry, primary metal; source, neutron; spectrometry; plant scale; Si, ^{238}Pu.*

Abstract: *A neutron activation-analysis system is described for the on-line measurement of silica in iron-ore slurries. The system is currently being used 24 hr/day in Canadian and U. S. iron-ore processing plants. A ^{238}Pu–Be neutron source is used to activate the silica in the iron-ore slurry, which is continuously recirculated between the source and a NaI(Tl) detector. The system is automatically compensated for variation of the solids content of the slurry by means of a gamma-ray transmission gage. The analyzer may be coupled to a computer for rapid feedback of analysis results to the plant process-control system, or the percent silica can be printed out on paper tape or provided as an analog signal. The standard deviation between neutron activation and wet-chemistry silica determinations was ±0.10% silica in the concentration range 3 to 11% silica.*

Activation analysis had its beginnings in the work of Hevesy and Levi[1] in 1936 and of Seaborg and Livingood[2] in 1938. Since then many technical innovations have been made so that today the method is highly competitive with other analytical techniques. The relatively high penetration of neutrons and of emitted gamma rays makes activation analysis a superior technique for measurement of low-atomic-number elements in bulk materials. No special thin-sample presentation cells or windows are required. The great depth of penetration minimizes errors due to heterogeneity in analysis of coarse solids in slurries with variable particle size. The weight of the material being measured, whether a continuous or batch sample, can often be made large enough to be representative of the heterogeneous materials in the process stream at that time, thus removing a very important source of error. Undoubtedly the greatest potential of activation analysis is in the area of industrial process control, as is now being recognized in the mineral-processing industry. Key elements in a process stream can be determined rapidly and accurately for immediate feedback to the process-control system. With the recent availability of inexpensive radioisotopic sources of medium-to-high neutron intensity and of reliable, long-life, sealed-tube neutron generators, activation analysis is now becoming economically attractive as an analytical tool in process control.

This article describes NOLA I‡ (Neutron On-Line Analyzer), a completely automated activation-analysis system for the measurement of silica in iron-ore slurries which uses a radioisotope neutron source. A ^{238}Pu–Be neutron source is used to activate an iron-ore slurry, which is continuously recirculated between the source and a NaI(Tl) detector. Automatic compensation for variable solids content in the slurry is achieved with a gamma-ray transmission gage. In a measurement time of 6 min, for a 50% solids sample, the measured standard deviation between NOLA I and wet chemistry is ±0.10% silica. Slurries containing less solids or shorter analysis times can be accommodated with some sacrifice in relative accuracy.

NOLA I is currently being used 24 hr/day in Canadian and U. S. taconite-processing plants. To our knowledge, these are the first industrial systems installed in the United States for slurry-activation analysis using a radioisotope source. This instrument offers the advantage of speed, convenience, and automation of earlier X-ray systems, but performs the analysis independent of heterogeneity, particle size, or chemical matrix effects.

TECHNIQUE

When the sample to be analyzed is in the form of a flowing solution or slurry, the sample may be handled in either a *flow-through* mode, where the sample stream passes the irradiator and detector only once during the analysis cycle, or in a *recirculation* mode, in

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†Texas Nuclear Division, Nuclear-Chicago Corporation, Austin, Tex. The contributions of many colleagues and helpful cooperation of the staff of Pickands Mather & Co.–Erie Mining in the development of this instrument are gratefully acknowledged. Special appreciation is due W. H. Tuttle and G. A. Peterson for supplying the wet-chemistry data presented in Fig. 4.

‡Trademark of Nuclear-Chicago Corporation.

which a portion of the sample stream is drawn off and continuously recirculated between the irradiator and detector during the analysis cycle. An in-depth comparison of these two analysis modes is given by Ashe, Berry, and Rhodes[3] and Berry and Martin.[4]

Recirculation analysis is used in NOLA I for two reasons. First, the sensitivity can be made independent of stream flow-rate variations over a wide range by careful design of the sample-loop variables. Flow-rate variations cause large fluctuations in the sensitivity of a flow-through system and a corresponding decrease in the accuracy of silica measurement. Second, a recirculation system can provide a higher sensitivity than a flow-through system with the same sample-loop design. The magnitude of this increase depends on the half-life of the induced radionuclide, the sample residence time in the several components of the loop, and the point, in the time available for analysis, at which counting is begun. In the NOLA I silica analyzer, for example, the recirculation mode is approximately 12 times as sensitive as the flow-through mode if counting is begun after radioactive equilibrium is attained. However, NOLA I is designed for use in process-control applications where the analysis time must be held to a minimum; so counting is begun immediately after the sample is injected into the loop—i.e., several minutes before equilibrium is attained. In this case, the recirculation mode is still approximately six times as sensitive as the flow-through mode.

SYSTEM DESCRIPTION

A schematic of NOLA I is shown in Fig. 1. Before each analysis, the system is in a standby condition with water recirculating in the analysis loop. When a sample is presented for analysis, a two-way valve introduces the slurry into the loop and ejects the water. When the loop is fully charged with slurry, the two-way valve is returned to the recirculation position, and measurement of the induced activity at the detector and the transmitted gamma rays at the density gage is begun and continued for a preset time. At the end of this period, data from the detector and density gage are output either to a printer or to a computer for interpretation. At the same time, the two-way valve is actuated to flush the loop with water in preparation

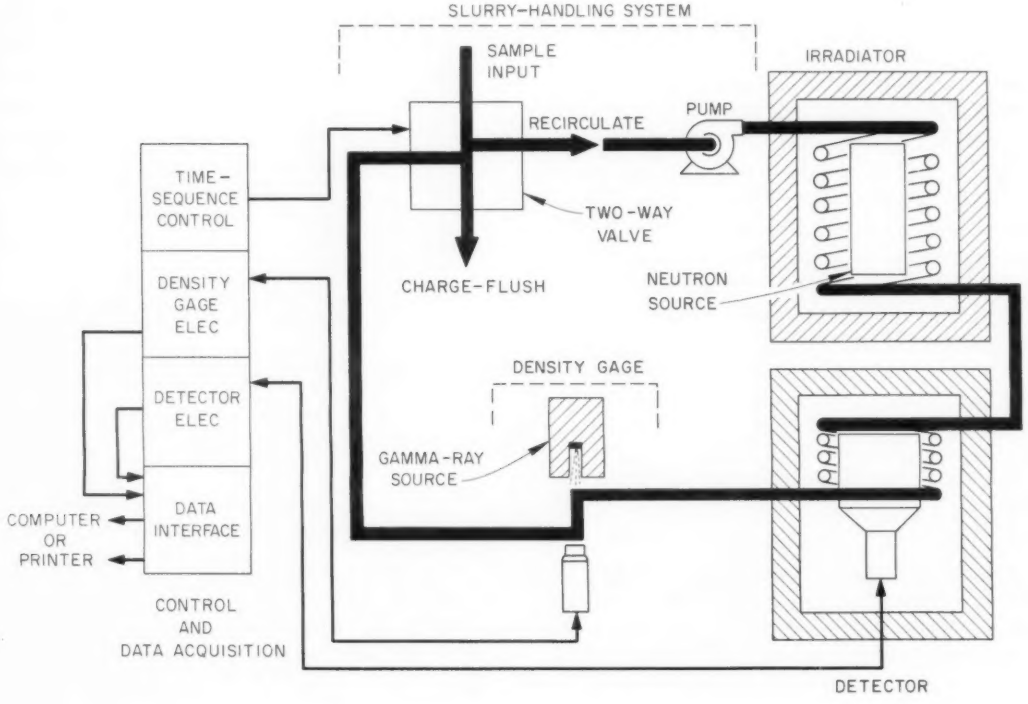

Fig. 1 Schematic representation of NOLA I.

for the next sample. The entire analysis cycle, including charging and flushing of the loop, is performed within 6 min.

From an operational viewpoint, NOLA I may be divided into five basic subsystems: slurry-handling system, irradiator, detector, density gage, and control and data-acquisition system (Fig. 1).

The slurry-handling system consists of holding tank, slurry agitator, two-way valve, and analysis loop. The paths taken by the sample in the analysis loop for each of the two valve positions are shown in Fig. 1. In the charge–flush mode, the analysis loop is open on both ends, one end being the sample intake from the holding tank and the other being the outlet for the analyzed sample or flush water. In the recirculation mode of the two-way valve, the analysis loop is a closed system.

The NOLA I irradiator is a ^{238}Pu–Be neutron source, a fully encapsulated activation cell, an oil-filled inner source shield, and a water-filled outer source shield. The neutron source has a nominal yield of 10^8 n/sec with a half-life of 87.6 years. The activation cell, in which the sample is exposed to the neutron flux, is fully encapsulated; it may be removed from the inner shield without disturbing the neutron source. The outer source shield is water filled and has a level sensor and automatic fill to compensate for evaporation.

For the determination of silicon, neutrons with energies greater than 3.86 MeV interact with the nuclide ^{28}Si in naturally occurring silicon to produce ^{28}Al:

$$^{28}\mathrm{Si} + n \rightarrow {}^{28}\mathrm{Al} + p$$

The ^{28}Al thus formed is an unstable nuclide and reverts to the original stable nuclide ^{28}Si by emitting a beta particle accompanied by a 1.78-MeV gamma ray:

$$^{28}\mathrm{Al} \rightarrow {}^{28}\mathrm{Si} + \beta + \gamma\,(1.78\ \mathrm{MeV})$$

The half-life of the decay is 2.25 min.

From the irradiator, the activated sample moves to the count cell in the detector where the induced ^{28}Al activity is measured. A schematic of the detector (Fig. 2) shows the detector electronics and a typical pulse-height spectrum from an activated iron-ore slurry. A NaI(Tl) crystal is used to measure the induced radioactivity from the sample. Pulses corresponding to the photopeak and part of the Compton tail from the 1.78-MeV gamma rays from ^{28}Al are isolated with a single-channel analyzer. The overall system gain is stabilized by use of a reference peak from an ^{241}Am alpha-particle source embedded in the face of the NaI(Tl) crystal. The stabilizer rapidly samples the count rate from each side of the peak and maintains

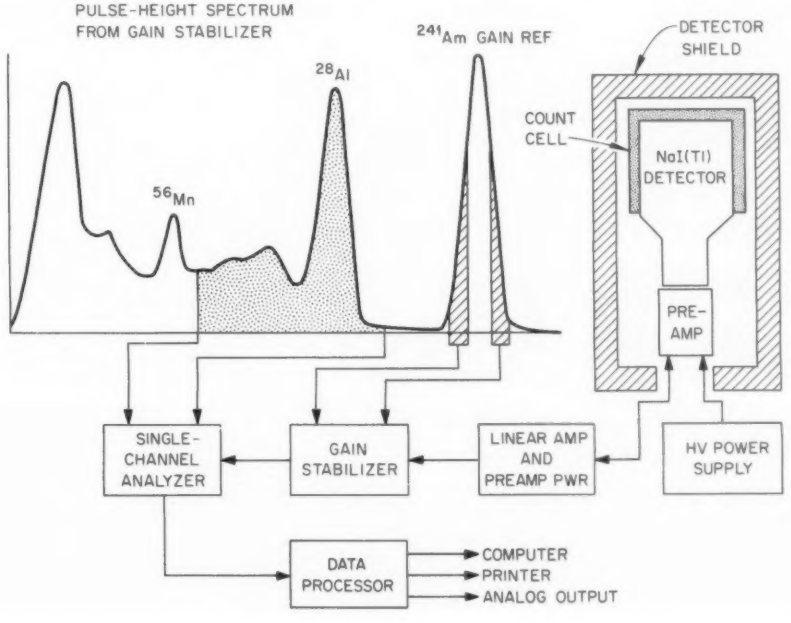

Fig. 2 Schematic of NOLA I detector showing typical gamma-ray spectrum from an activated iron-ore slurry.

the system gain at the point at which the two count rates are equal.

Two other reactions may interfere in the measurement of silicon in iron ore. The radionuclide ^{56}Mn, with a half-life of 2.58 hr, is produced by the ^{56}Fe(n,p) reaction. This decay results in gamma rays of 0.845, 1.81, and 2.13 MeV. The 0.845-MeV gamma ray is shown in the pulse-height spectrum of Fig. 2; however, the 1.81- and 2.13-MeV lines are masked by the 1.78-MeV line from ^{28}Al. The 0.845-MeV line is eliminated with the single-channel analyzer. The interference from the two higher energy lines is rendered insignificant in NOLA I by proper selection of the length of activation time. The other likely source of interference is from the ^{27}Al(n,γ)^{28}Al reaction. Since the end product of this reaction is the same as from the ^{28}Si(n,p) reaction used in the measurement, there is no way to eliminate this interference electronically. However, the interference can be reduced to a negligible level by proper selection of the activation-cell volume and by surrounding the activation cell with a cadmium shield.

NOLA I specifically measures the mass of silicon present in the activation loop in each analysis sequence. The quantity of interest for process control is the percent by weight of silica in the iron ore. Because the solids content of the iron-ore slurries being analyzed may vary from 30 to 60%, an additional measurement of the solids mass in the analysis loop must be made to obtain percent by weight of the element. In NOLA I this is accomplished by a density gage consisting of a NaI(Tl) detector and a Nuclear-Chicago model 5176 density-gage head that provides a collimated beam of 0.662-MeV gamma rays from a ^{137}Cs radioisotope source. The intensity of the gamma-ray beam transmitted through a segment of the slurry path is measured with the NaI(Tl) detector and used to normalize the silicon signal to a given solids mass (50%) in the loop. The percent by weight silica in the loop may then be computed from these two measurements, if all the silicon in the sample is assumed to be present as silica.

The physical configuration of the NOLA I silica analyzer is shown in Fig. 3. The control and data-

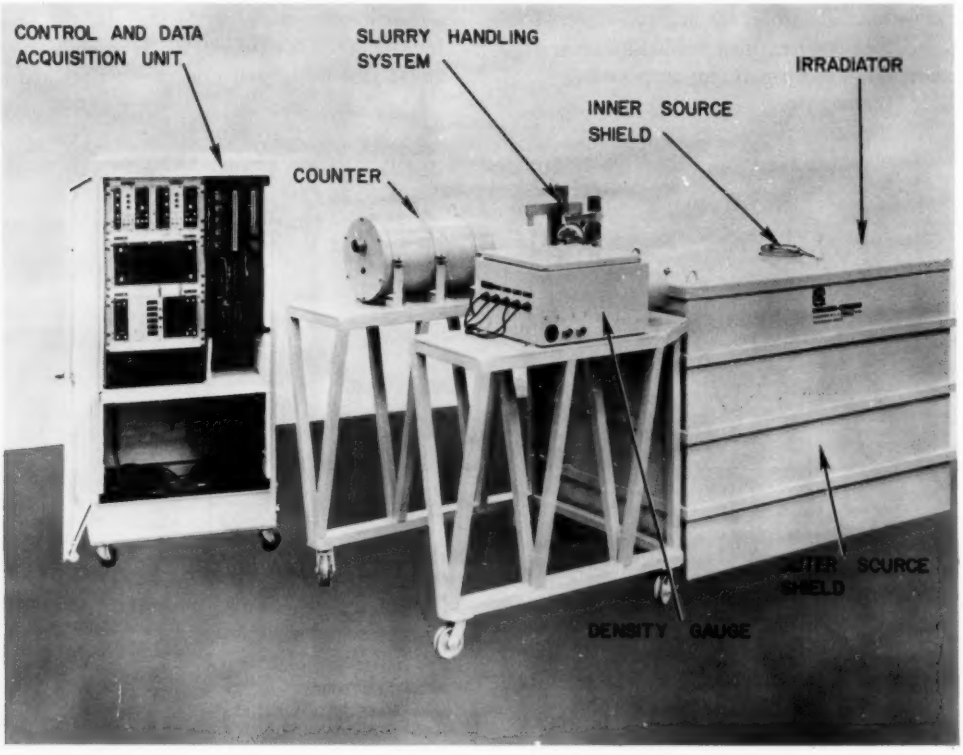

Fig. 3 NOLA I silica analyzer.

acquisition unit houses the auxiliary electronics for the two detectors, a time-sequence controller, and a data analyzer. Three output options are provided: (1) a digital record of the percent weight silica; time and date can be printed locally or at a remote station, (2) a digital signal that can be interfaced into a computer, and (3) an analog signal, 0 to 10 V or 4 to 20 mA, that can be used for process control. The control and data-acquisition system may be completely contained in a controlled-environment cabinet for operation under particularly harsh conditions; however, in most cases, forced-air cooling is satisfactory.

INPUT REQUIREMENTS

The NOLA I system is designed so that it can be used in the analytical laboratory to analyze batch samples, or on the plant floor as a continuous process monitor. In the analytical laboratory samples can be added manually to the slurry-holding tank. The measurement cycle is then initiated, and after analysis a printed record of the time, date, and silica content of the sample is provided. As a process monitor on the plant floor, samples can be cycled automatically into the holding tank from various locations within the plant. Again, printed records can be obtained, or data may be interfaced into the plant computer or the analog signal may be used for process control. Monitoring the input and output of the beneficiation circuit would allow continuous optimization of this equipment whether it be a grinding mill, magnetic separator, or flotation cell. The sample size required, 1 liter in the holding tank, is large enough to be representative of the process stream but small enough to handle easily. The volume of the slurry loop is approximately 500 ml, and the solids content can range from 30 to 50% solids. The slurry-handling system can accommodate material with a maximum particle size of 60 mesh.

After analysis, the residual activity of the sample is so low and short lived that the material can be returned to the process stream.

ECONOMIC JUSTIFICATION

Silica-content specifications for the taconite pellets are generally established by the end user—the blast furnace operator. These specifications are established in order to permit accurate prediction of the physical characteristics of the melt and eliminate the costly recycling of the material caused by erroneous blending. The advantages of rapid silica analysis are apparent in the ore beneficiation stage, when the pellet producer requires the information to maintain constant grade and is faced with continuing pressure to reduce the silica level. Instrumental control of the beneficiation circuit, whether it be a grinding mill, magnetic separator, or flotation cell provides a superior product at lower cost relative to hand-controlled operation. In addition, the energy needed to grind the ore can be optimized since the grinding required is a function of the ore's silica content. However, economic savings are difficult to calculate in this situation where premiums are not paid for the lower silica-content pellets, and the major difficulty is encountered in marketing the product.

Cost per silica analysis, using NOLA I, compared to wet chemistry is more readily obtained. The price of a NOLA I instrument, its initial installation, and its startup cost comprise a total investment that would be depreciated over a 5-year period. Assuming the operation efficiency would be 90%, and including labor costs to perform periodic maintenance, the total cost per analysis would be less than $0.45. Hence, the NOLA I unit can perform silica determinations on-line, at a cost of approximately one-fifth that of commercial analytical services, on a continuing basis and eliminates many of the errors that occur in routine wet-chemistry techniques.

SYSTEM PERFORMANCE

NOLA I has been extensively tested both in the laboratory and after installation in the field. The first NOLA I installation has now been operating satisfactorily 24 hr/day in an iron-ore processing plant for approximately 12 months. Calibration curves such as the one shown in Fig. 4 have been obtained with this

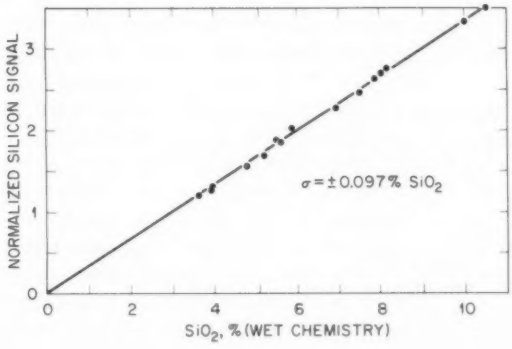

Fig. 4 Typical NOLA I calibration curve for analysis of silica in iron ore.

instrument. The background-corrected silicon counts, normalized to 50% solids slurry by the gamma-ray transmission-gage measurement, are presented on the ordinate vs. the percent by weight silica determined by wet-chemistry techniques on the abscissa. A least-squares analysis applied to these data determined the best straight-line fit. The standard deviation about the line is ±0.1% silica. The system precision for repeated measurements on identical samples was ±0.06% silica for a sample with nominal 6% silica concentration. Many other samples from widely separated locations in the United States and Canada have been evaluated with similar results.

CONCLUSIONS

Neutron activation analysis has been found to be an accurate, rapid technique for measurement of silicon in iron-ore process streams. The acceptance of NOLA I by the iron-ore processing industry as a routine process-control tool should pave the way for the routine use of activation analysis in other industries. There are several elements—including aluminum, copper, fluorine, and potassium—important in process-control and pollution-control applications which appear amenable to accurate, rapid determination by on-line neutron activation-analysis techniques. (RHL)

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BRITISH BOREHOLE PROBE ASSAYS TIN ORE UNDERGROUND

A borehole probe, developed under UKAEA sponsorship, for the in situ assay of ore containing 0.1% or more tin is about to come into commercial production in Britain by Ekco Instruments Ltd., Southend-on-Sea, Essex. The instrument is based on nondispersive X-ray-fluorescence spectrometry with a radioactive source. It is intended to meet the needs of the mining industry for a quick and inexpensive way of assessing the commercial value of underground ore bodies, particularly those near to, or approachable from, existing workings, without the necessity for taking samples or for extensive sample preparation or analysis. Results of field tests in horizontal boreholes in the recently developed Wheal Jane mine in Cornwall agreed closely with chemical analyses of cores from the same borehole.

The entire equipment is transportable, both on the surface and underground, and comprises a borehole probe connected by 50 ft of flexible cable to a light trolley that carries the control and the electronic readout unit, the battery power pack, and a compressed-air cylinder. The probe is a 24- by 1.75-in. stainless-steel cylinder, housing an annular radioactive source, a pair of balanced X-ray filters (operated pneumatically), and a plastic scintillator with photomultiplier. The probe will operate in boreholes of $1\frac{15}{16}$ and $2\frac{1}{4}$ in. diameter under wet or dry conditions, and it can tolerate substantial unevenness in the borehole walls. A calibrated flexible hose carrying the air tubes for the filter actuator and the electric power and signal cable to the surface also accurately controls and indicates the location of the probe in the hole. The electronic readout is in digital form and indicates tin concentrations of 0.1% and more in a measuring time of <0.5 min for each position of the probe. For comparison, the traditional method of taking borehole cores and analyzing them chemically takes a number of days. (MG)

Energy-Dispersion X-Ray (EDX) Analysis in the Nonferrous Mining Industry*

By A. P. Langheinrich, J. W. Forster, and T. A. Linn, Jr.†

Supplementary Keywords: *X-ray fluorescence; industry, mining; metallurgy, nonferrous; spectrometry; Cu; Fe; Mo; Pb; Zn; U; ^{57}Co; ^{109}Cd; ^{125}I; ^{238}Pu; ^{241}Am.*

Abstract: *Energy-dispersion X-ray (EDX) spectrometry based on Si(Li) detectors is approximately 5 years old. Although its first application in the mineral industries was reported in 1967, it is only now being accepted for its unique capabilities and considered competitive with other analytical techniques.*

Energy-dispersion X-ray equipment in combination with radioisotope sources was used successfully in the analysis of mining and milling samples for Cu, Fe, Mo, Pb, Zn, and U. Sample types included solids, solutions, muds, and slurries. A variety of modular combinations of sources, sample cells, Si(Li) detectors, amplifiers, single- and multichannel analyzers, scalers, rate meters, printers, and recorders was used to fit particular jobs. Test results and field applications have demonstrated the value of EDX instrumentation as an analytical tool. Its future wide use in laboratory, pilot plant, and process plant is expected.

Single-crystal semiconductor detectors [lithium-drifted silicon, Si(Li)] were first used in 1966 for the detection and energy separation of characteristic X-ray spectrums.[1-3] Although energy-dispersive X-ray (EDX) detection was not new at that time, Si(Li) detectors in combination with pulse-height analyzers provided the degree of energy resolution that encouraged practical applications and resulted in instrumentation competitive with conventional wavelength-dispersive apparatus. Because of high detection efficiencies, such spectrometers did not require intense X-ray excitation sources. Small radioisotope sources could be used in place of the usual high-voltage X-ray generator.

Since 1966, detector resolution for a fixed detector area has been steadily improved; while a resolution of 650 eV was remarkable then, resolutions of better than 200 eV are now common. Sufficient resolution and the high counting abilities of large-area detectors today allow rapid and routine analysis of many chemical elements, even of neighboring atomic number, over wide ranges of concentrations in a great variety of materials.

Although many applications of radioisotope X-ray techniques in the nonferrous mining industry have been reported,[4-12] reports on the combination of radioisotope excitation and Si(Li) detection are few.[13-15] Within Kennecott Copper Corp., the following EDX work has been done since 1966 or has been requested by operating divisions for the near future: copper and iron determinations in mine-water solutions, leach liquors, ore and waste materials, and mill headings, tailings, concentrates, and intermediates; copper in electrolytes; lead, zinc, silver, and cadmium in Pb–Zn ores, concentrates, and process intermediates; uranium in aqueous and organic solutions and in solid products; and molybdenum in copper and molybdenum circuit materials and molybdenum mill-heading and -tailing samples. Many of these analyses were requested for situations where quicker answers than are currently available would result in better process control and, hence, in savings.

In addition to its semiquantitative and quantitative capabilities, EDX analysis is a powerful qualitative tool. Furthermore, Si(Li) detectors have found wide acceptance in microprobe spectroscopy[16] and in miniature probe work.[17]

This article discusses the equipment utilized at Kennecott Research and presents examples of applications and results.

EQUIPMENT

The instrumentation for EDX analysis in metal mining and processing had to be reliable, simple to use, and sufficiently flexible to meet the economic and analytical requirements of specific projects. A typical system for single-element analysis was assembled in the

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†Kennecott Copper Corp., Metal Mining Division, Research Department, P. O. Box 11299, Salt Lake City, Utah 84111. The authors thank the Kennecott Copper Corp. for permission to publish this paper and acknowledge the interest in this development work as well as suggestions and encouragement of H. R. Spedden, Research Director, and W. M. Tuddenham, Section Chief.

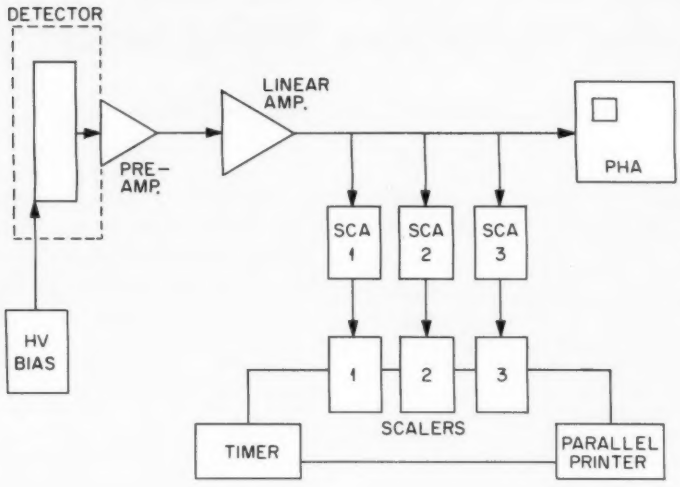

Fig. 1 Three-channel EDX system (AMP. = amplifier, PHA = pulse-height analyzer, SCA = single-channel analyzer).

laboratory for industrial application, specifically for uranium determinations. It includes a sample–source–detector assembly (including preamplifier), linear amplifier, single-channel analyzer, and a nuclear counter with crystal time base. The digital display provides an integrated count that is related by a calibration curve to the concentration of uranium in the sample. Energy calibration is accomplished by the use of a calibration standard and proper adjustment of the single-channel analyzer.

The block diagram of a basic EDX analyzer for the determination of three elements is shown in Fig. 1. Fluorescent X rays from the sample are excited by a small radioisotope source in a holder attached to the detector. Each single-channel analyzer drives a scaler for integral counting and data accumulation. The multichannel analyzer is used in the initial system calibration for adjusting the amplifier gain and the single-channel analyzer windows. Systems such as this one can be assembled for less than $15,000, including radioisotope sources.

The compactness of a typical EDX system is shown in Fig. 2. This particular unit is mounted in a trailer for use in the field.

Fig. 2 Trailer-mounted EDX field laboratory.

Excitation Sources and Sample Cells

Both low-power conventional X-ray tubes and radioisotope sources have been used successfully in this

work. The radiation properties of several isotopes are summarized in Fig. 3, with the useful range of each source for K-series and L-series X-ray excitation of the chemical elements. Reasonable coverage for the elements from calcium through uranium can be obtained with proper selection of the excitation source. The problem of source decay is insignificant for plutonium and americium sources. For sources with shorter

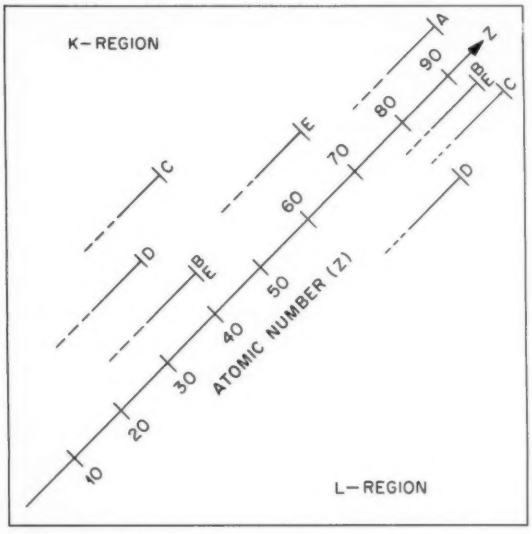

Fig. 3 Radioisotope source data. A, 270-day ^{57}Co; 14-, 122-, and 136-keV gamma rays; 6.4-keV iron K X rays. B, 453-day ^{109}Cd; 22-keV silver K X rays. C, 60.2-day ^{125}I; 27-keV tellurium K X rays. D, 86.4-year ^{238}Pu; 12- to 17-keV uranium L X rays. E, 458-year ^{241}Am; 11- to 22-keV neptunium L X rays; 60-keV gamma rays.

half-lives, permanent standards have been prepared containing varying amounts of the element of interest embedded in Lucite disks. These plastic standards are counted in the same manner as the primary standards from which the calibration curve was constructed. By use of the calibration curve, concentration values can be assigned to these plastic disks, and the assigned concentration values plus X-ray counts allow updating of calibration curves whenever decay corrections are necessary. Because of the short half-life of ^{125}I, ^{109}Cd has been substituted, particularly for molybdenum analysis. Both ^{109}Cd and ^{238}Pu sources are sealed in stainless-steel capsules with aluminum windows. A source holder with three 30-mCi ^{238}Pu sources is shown in Fig. 4 together with a disassembled sample holder for solids and wet pulp. Similar source-holder

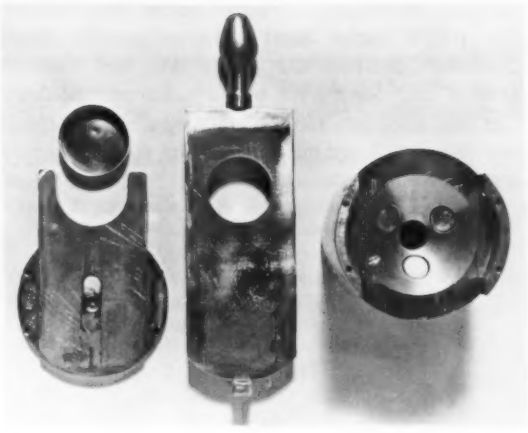

Fig. 4 Source and sample-holder components for dry and wet samples.

construction is used for the ^{109}Cd sources. The aluminum holders, designed and fabricated in the laboratory machine shop, provide adequate shielding of the radioisotopes to prevent personnel exposure even over long periods of operation.

The source and sample-holder design evolved from the use of several different source–sample configurations and was based primarily on geometric analysis of the physical problems associated with irradiating the sample and collecting maximum counts. (For the source configuration shown in Fig. 4, a maximum count rate was obtained 0.5 in. from the source to the sample surface.)

The sample cell for liquids has been used in the laboratory for many years and has saved countless hours of operator time when compared with single-cell preparation methods for solution samples. Both aqueous and nonaqueous solutions are readily accommodated. The cell geometry and capacity were designed so that memory effects are practically eliminated. A similar cell has been used for on-stream analysis of solutions and slurries. A typical detector–source–sample arrangement is shown in Fig. 5.

Detectors

Good Si(Li) detectors are available from a number of manufacturers, and resolutions of better than 200 eV are now commonplace. Resolution alone, however, is not a decisive factor in industrial applications. Although high resolution is desirable in electron microprobe work and in the determination of low-atomic-number elements, a resolution of 400 eV was

satisfactory in most of the applications described here. The proper choice must be a compromise among resolution, detector size, count rate, and allowable analysis time for a given problem. Commercial detectors have good long-term stability even when thermally recycled, and liquid-nitrogen cooling was less troublesome than anticipated.

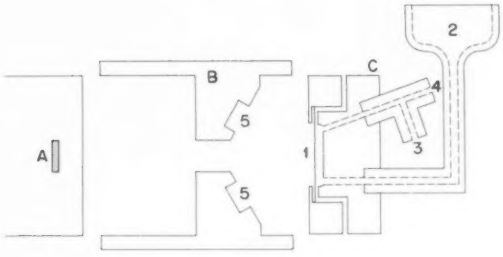

Fig. 5 Exploded view of typical detector (A) source holder, (B) solution sample holder, (C) arrangement [(1) Mylar window, (2) sample inlet, (3) sample discard, (4) air outlet and level control, (5) source locations].

Nuclear Instrument Modules (NIM)

The NIM system assured the required flexibility for this work. After initial testing with a multichannel analyzer, single-channel analyzers have been substituted in all field applications. Because of the high quality and stability of the modules used, pulsers, analog stabilizers, and base-line restorers in industrial units have been unnecessary—with accompanying cost reductions. Another significant advantage of the NIM system is the ready interchangeability of modules. In event of breakdown, modules can be interchanged by operating personnel. This results in very short downtimes. Because of the relatively low cost of the modules, spare units can be stored centrally for rapid replacement where necessary. Such systems can be expanded easily to accommodate additional channels for determination of additional elements. Space required is minimal, and the only power required is a stabilized 110-V line. For single-element systems, economics dictates the use of display scalers; but, for reasons of speed and convenience, digital printers are preferred in multielement analysis. For on-stream work, a recycle timed readout or a combination rate meter–strip-chart recorder for continuous analysis can be selected. The system can be connected to a central data processor or computer for direct process control.

The advantages of radioisotopic EDX equipment are: (1) no optics, goniometer, or moving parts; (2) no spectral-order interferences; (3) no X-ray tube, power supply, or associated heavy shielding; (4) lesser matrix and particle-size effects; (5) small size, low cost, and portability; and (6) excitation of K-series X rays of high-Z elements. Disadvantages of the method include: (1) need for liquid-nitrogen cooling; (2) limited counting rates and longer counting times; (3) need for multiple sources; and (4) source decay of short-half-life radioisotopes. While ordinary X-ray-fluorescence equipment has advantages in certain cases, EDX apparatus is generally attractive for many applications in the nonferrous mining industry.

APPLICATIONS

Recently EDX analysis has developed into an accepted routine analytical tool, and its use is expanding at an increasing rate. The kinds of materials analyzed in the past have included mining and mill samples and test samples from various research projects. The physical condition of such samples varied widely. Most of this work was done with horizontal-detector systems. This configuration is adequate for solutions, slurries, and dry solids, but it is unsatisfactory for wet, muddy samples, which require a vertical or angular detector configuration.

Molybdenum Assay

Heading and tailing samples from a molybdenum mill were investigated by EDX analysis. Such analyses can be conducted on the operating floor of the mill, and results are available within about 5 min. About 80 samples can be analyzed in an 8-hr shift by a mill floor operator. Samples were dried, ground, and pressed at 20,000 psi into $1\frac{1}{8}$-in.-diameter aluminum cups. A comparison of chemical assays and the results of EDX analysis is given in Fig. 6. Point deviations from the least-squares line are indicated by the standard-error value. The EDX results compare well with conventional X-ray data and are within the reproducibility of chemical assays for these samples. The radioisotope source used in this test was relatively weak—^{125}I with an approximate activity of only 0.9 mCi. Since the half-life of ^{125}I is only 60 days, decay corrections had to be made periodically with this source. Other work on molybdenum has been reported previously.[13]

Copper and Iron Assay

Figure 7 illustrates the results obtained from EDX analysis of samples from copper-milling operations. The purpose of this study was to establish the

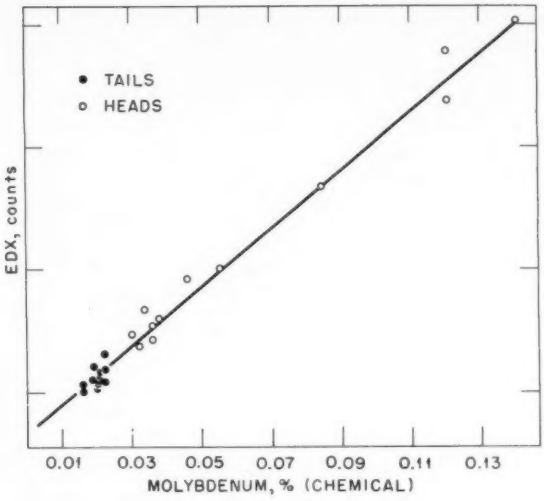

Fig. 6 Molybdenum in mill samples. Chemical reproducibility S.D. = 0.0067; conventional X-ray S.E. = 0.0032; EDX S.E. = 0.0032. Iodine-125 source 0.9 mCi.

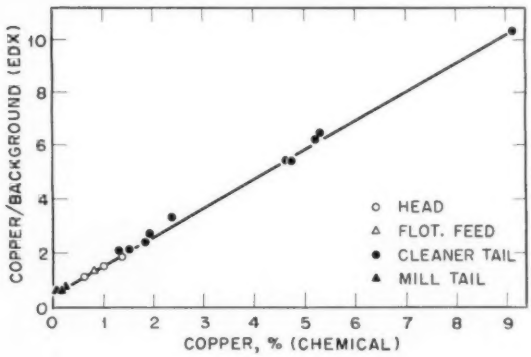

Fig. 7 EDX analysis of dried, pressed mill samples. Plutonium-238 source 20 mCi. S.E. = 0.13.

equivalence of the EDX method with conventional techniques so that recommendations for in-plant and on-site uses could be made. The samples were analyzed both as received from the mill and after regrinding. Statistical evaluation of the results (Table 1) showed that regrinding did not improve the standard error of estimate—a conclusion reached on a number of previous tests but not yet fully understood. A significant improvement in the data was noted when the copper counts were corrected using a ratio with the uranium L-alpha X-rays from the ^{238}Pu source. Figure 8 illustrates this application of EDX to the assay of

Table 1 Statistical Data for Copper in Mill Samples (Range 0.15 to 9.13% Cu)

Method	Sample condition	Standard error of measurements
Crystal dispersion Cu	As received	0.41
Crystal dispersion Cu/R_1 *	As received	0.12
Crystal dispersion Cu	Reground	0.51
Crystal dispersion Cu/R_1 *	Reground	0.19
Energy dispersion Cu	As received	0.49
Energy dispersion Cu/R_2 †	As received	0.13
Energy dispersion Cu	Reground	0.67
Energy dispersion Cu/R_2 †	Reground	0.26

*R_1 = 29.00° (2θ), LiF = 1.01 Å.
†R_2 = $U_{L\alpha}$ peak from ^{238}Pu source.

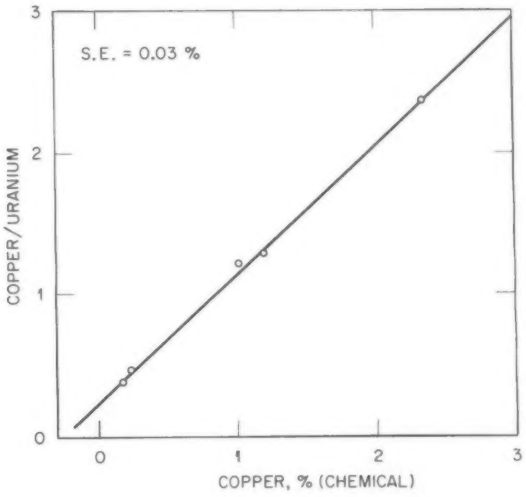

Fig. 8 EDX analysis of wet mill samples. Plutonium-238 excitation, 4-min counts.

copper in wet mill samples and gives a comparison with chemical assays of the same samples.

Field Analyses

Figure 9 shows calibration curves from a small field laboratory for the measurement of copper and iron in mine-water solutions.

Because of its speed and reliability, EDX analysis has replaced atomic-absorption spectroscopy in this field operation. The EDX system has been in use continuously for over 12 months with only minor maintenance problems even under extremely adverse environmental conditions. The system is readily con-

vertible to handle solid samples and both static and on-stream solutions. Three 30-mCi sources of ^{238}Pu are used. Standard samples are run on a regular basis to check for any malfunction of the equipment. Six hundred determinations are made each day. Especially in this project has EDX become established as a routine analytical tool.

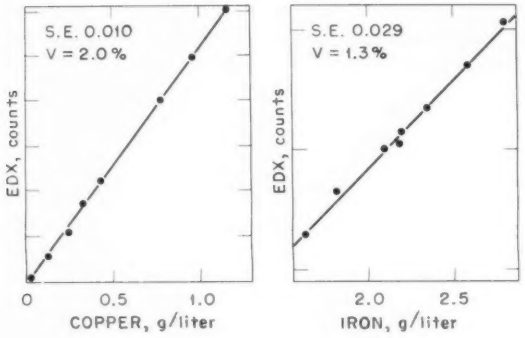

Fig. 9 Calibration curves for the routine EDX analysis of mine-water solutions.

The relative compactness and portability of basic EDX equipment suggested possible field applications, particularly ore and waste analyses at mining sites. A small detector head with cooling reservoir (Fig. 10) was built by an instrument vendor at our request. It has been used for in situ analyses inside vertical drill holes 25 ft deep and for the analysis of drill chips from drilling operations. Such analyses can provide rapid on-site information for mining personnel who must decide on shipment of the mineral-bearing rock either to processing plants or waste dumps. Typical chip samples (Fig. 11) were analyzed with a horizontally oriented detector. The sample was held in the sample cups with Mylar film fastened by rubber bands. Five chips from each of four different holes were investigated. After EDX determinations, the five samples from each hole were combined, particle analyzed, ground, and then chemically analyzed. Particle-size analysis showed +10 mesh fractions in the four holes of 12, 55, 19, and 33%, respectively. The chemical values were plotted vs. average EDX counts, and calibration curves were calculated from the data for copper and iron. The concentration range for copper for the four holes was 0.23 to 2.00% and for iron 5.20 to 19.0%. These calibration curves were used in the analysis of four unknown chip samples. Again EDX readings were taken on each of five subsamples. The average count

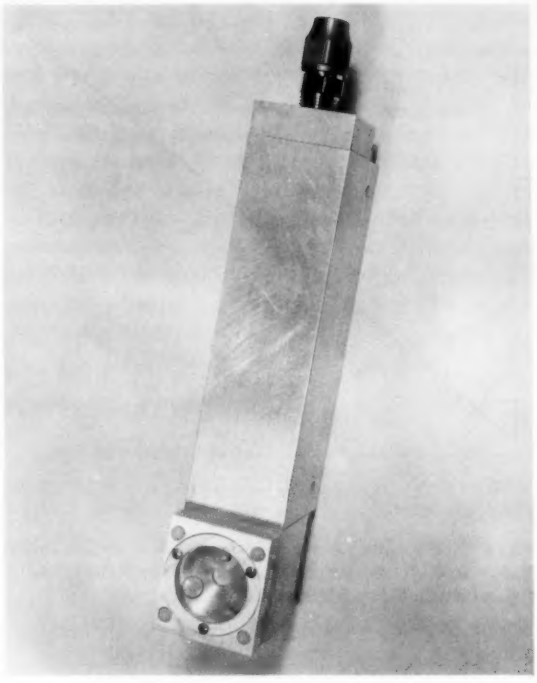

Fig. 10 EDX drill-hole probe.

Fig. 11 Drill-hole chip samples.

was used to determine copper and iron values. Results of the test are summarized in Table 2. The wide ranges for the subsamples are largely caused by true concentration differences. Repeat analyses on several samples by repacking and recounting the same material in the same cup five times gave standard deviations that averaged less than 0.2% for copper and 1.7% for iron. The test results led to the current practice of analyzing and averaging 5 to 10 samples above ground or integrating counts over the drill-hole depth.

Table 2 Comparison of EDX Analysis of Rock Chips* with Chemical-Assay Data for Copper and Iron Content

Sample	Number of determinations	Copper			Iron		
		Range	Mean, % Cu	Chemical assay, % Cu	Range	Mean, % Fe	Chemical assay, % Fe
141-A	5	1.21 to 1.60	1.45	1.70	19.3 to 21.0	20.2	19.1
141-B	5	1.40 to 2.97	1.89	1.64	14.2 to 16.5	15.4	14.4
142-A	5	0.42 to 0.54	0.49	0.46	12.4 to 13.9	12.7	11.2
142-B	5	0.07 to 0.60	0.23	0.18	4.8 to 5.9	5.8	4.5

*Rock chips taken from mine drill holes.

Another mine application was the analysis of dried, ground, and pressed samples of varying mineralogical/petrological composition. In spite of wide compositional variations, remarkable linearity was achieved (Fig. 12). This again points toward EDX as a convenient tool for ore and waste sorting.

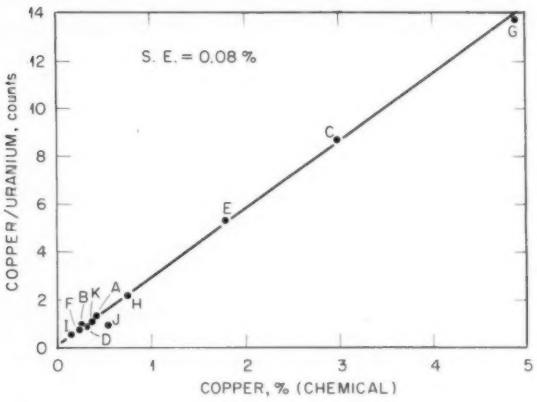

Fig. 12 EDX analysis of mine samples. Plutonium-238 excitation, 4-min counts. Medium iron: B, schist, cuprite; C, schist, chrysocolla, cuprite; J, schist, cuprite, natural copper. High iron: D, E, A, K, diabase, chalcopyrite. Low iron: F, porphyry, chalcopyrite; G, schist, chalcocite; H, schist, chalcocite; I, quartzite, chrysocolla.

Lead and Zinc Determination

A milling-plant application was the assay of lead and zinc (Fig. 13) in mill headings. All comparisons are based on single chemical-assay values. In lead concentrates, zinc rougher concentrates, lead cleaner tails, zinc cleaner tails, and rougher tails, the coefficient of variation for zinc data ranged from 0.6 to 4.4% depending on the particular material. These values are well within the requirements set by the operating personnel. As a consequence of such investigations, EDX equipment was introduced for long-term testing at a Kennecott-owned lead–zinc operation.

Uranium Determination

Another request for EDX work entailed the determination of uranium in solutions. Earlier, such determinations were carried out directly on samples of relatively high uranium concentrations and on solid materials including ion-exchange resins. For the rapid, simple, and quantitative determination of traces of uranium, however, a separation technique had to be applied. The separation was adapted from a procedure by Sill and Williams[18] which separates U(IV) and certain III- and IV-valent cations with barium sulfate in the presence of potassium. Uranium recoveries have

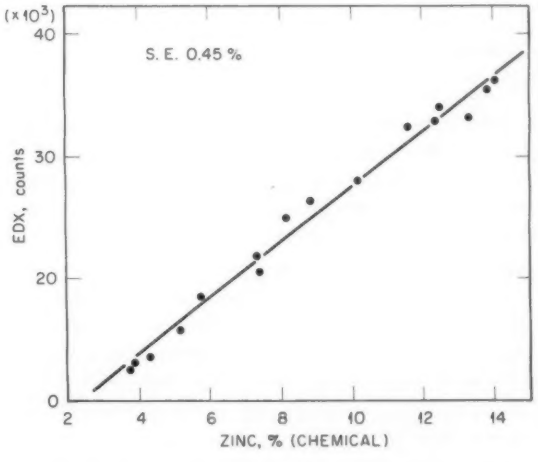

Fig. 13 Zinc in mill headings. Plutonium-238, 90-mCi excitation, 100-sec counts.

been better than 99.7% by this method. Following the separation, the uranium was determined by EDX analysis. The 22-keV silver X-rays from a ^{109}Cd source were used for the excitation of uranium L-alpha 13.6-keV X-rays. The single-channel analyzer was adjusted to discriminate against all pulses except those from the uranium L-alpha photopeak. Integrated counts of 100 sec were made. The method is used for aqueous samples containing from 5 to 100 μg of uranium in 1- to 25-ml aliquots. In extensive testing the relative precision was determined as 20% at the 1 ppM level and 4 to 7% at the 2 to 20 ppM uranium levels. Comparative assay results for uranium are shown in Table 3.

Table 3 Data Comparison for Uranium Assay (ppM U)

Sample	Method		
	Chemical*	EDX	Referee†
A	11	10	11
B	10	11	11
C	11	11	11
D	9	8	8
E	-	8.2	8.8
F	-	6.3	6.6
G	-	8.2	8.7
H	-	6.5	6.4
J	-	8.9	9.9
K	-	2.3	2.3

*Chemical: solvent extraction and spectrophotometric determination.

†Referee: analysis by U. S. Bureau of Mines, Salt Lake City (spectrophotometric).

SUMMARY

Because of its modular design, the EDX analysis system allows maximum flexibility in applications. Its size and portability are especially attractive for use in small field laboratories and at plant sites removed from the main analytical laboratory. The EDX system can provide data with precision equivalent to that of conventional X-ray analysis, with proper allowance for longer counting times in the case of EDX. The equipment assembled at Kennecott Research was used many months as the main instrument in the analytical support of plant operations and has been accepted by the operations personnel. The equipment has proven reliable both in the laboratory and in routine assignments for thousands of sample analyses. Recently an instrument was tested for several weeks by operations personnel and has passed this crucial test. From their final report, it can be concluded that EDX equipment has even greater application than originally proposed.

For the determination of iron and copper in process intermediates from several operating divisions, the use of a background correction or uranium peak ratio did not improve the analytical data for iron. For copper, however, use of the uranium peak ratio became necessary in most cases. In several tests, regrinding of a sample did not lower the statistical errors. Also, in most tests conducted, the results from wet (mud) samples were almost identical to those obtained from dried and pressed samples. The analysis of wet samples becomes important where quick answers are needed and time for thorough sample preparation is not available.

Presently five EDX systems are in use in laboratory research and in field installations for both testing and continuous operation in quality control. Developments in all areas discussed are continuing at an accelerated rate. A rapid growth of the technique can be expected, and the demand for instrumentation in numerous applications will challenge manufacturers to supply an adequate variety of laboratory and industrial EDX systems. (RHL)

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NUCLEAR TECHNIQUES IN ENVIRONMENTAL POLLUTION

Symposium Proceedings, Salzburg, Oct. 26–30, 1970, IAEA, Vienna, 1971
(STI/PUB/268) (CONF-701023).

The conference of which this is the proceedings was reported in *Isotopes and Radiation Technology*, 8(4). In the 810-page document, the 54 papers are grouped according to subject: nuclear techniques and pollution studies, determination of air pollutants, determination of water pollutants, mercury and other metal contaminants of the environment, movement of pollutants in water and soil, tracer techniques in coastal pollution studies, nuclear techniques and ecosystems, and sludge and waste-water treatment. Approximately 180 participants in the conference represented 33 countries—highly industrialized as well as developing—and 8 international organizations. The first paper in the proceedings, by W. E. Mott of the USAEC Division of Isotopes Development, reviews primarily work of the past 5 years in which nuclear techniques are applied. In his concluding remarks, Knut Ljunggren of Stockholm states that accurate and reliable analytical methods are essential for meaningful environmental studies. In between these two discussions are presented many of the current nuclear techniques of interest to persons concerned with environmental pollution. (MG)

*TECHNETIUM AS A CATALYST IN ORGANIC REACTIONS**

Supplementary Keywords: *catalysis; industry, chemical; industry, petroleum and natural gas; synthesis;* ^{99}Tc.

In preliminary test-tube- and bench-scale tests, technetium was shown to be potentially useful as a catalyst in the re-forming of petroleum fractions. For example, at 500°C and a contact time of 3 sec, *n*-heptane was 70% converted to a product that was 90% toluene. There was also indication of conversion of *n*-octane to ethyl benzene and *o*-xylene.

Of eight catalysts prepared, the best was one in which ammonium pertechnetate solution was adsorbed on column-dried chromatography-grade alumina (Fisher, 100 to 200 mesh) and reduced with H_2. The catalyst was readily regenerated with molecular oxygen at 500°C followed by reduction with hydrogen. Other technetium catalysts prepared on a different type of alumina or on Molecular Sieves were less satisfactory since with them the cracking activity predominated over the re-forming activity.

The product showed no radioactivity greater than background, indicating that no technetium was carried over. (MG)

*Summary of Final Report RLO-2017-3, by A. U. Beackham and J. C. Palmer, Department of Chemistry, Brigham Young University, Provo, Utah, Sept. 30, 1970.

Radioisotopic Monitor for Rime Accumulation

By W. B. Fowler*

Supplementary Keywords: *measurement, other; source, gamma;* ^{226}Ra.

Rime is a significant source of moisture in high-elevation forests, but weighing methods to determine the rate of its accumulation are tedious and difficult to apply due to high winds and constraining ice growth. Therefore a radioisotopic gage was constructed to measure rime accumulation under these adverse conditions. It was installed on a 6000-ft mountain in the eastern Washington Cascades, where it has given a continuous record of rime-accumulation data that agree well with weight data from nearby foliage. The apparatus operates continuously for 45 days without attendance.

The rime thickness at any given time is indicated by the attenuation of gamma radiation as it passes through the rime layer (Fig. 1). The radioactive source

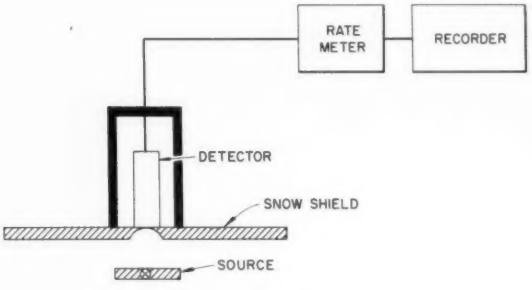

Fig. 1 Rime-accumulation-monitor schematic.

is 2 to 3 μg of ^{226}Ra encapsulated in plastic and located in a 10-cm-diameter by 1.5-cm-thick disk. Because the rime attachment to the smooth surface of the source disk was very tenuous and accumulations were often swept away by the wind, a 1-cm-thick low-density (0.03 mg/cm^3) hair pad was attached to the disk as a mechanical support for the growing deposit. The disk is suspended (Fig. 2) 15 cm beneath a detector—a halogen-quenched Geiger–Mueller tube with a mica end window—located inside a hermetic container that rests on a 60-cm-diameter 3-cm-thick snow shield. A recess in the snow shield just below the detector window prevents rime accumulation on the detector window. The count-rate meter showed readings of 850 counts/min when no rime was present.

(MG)

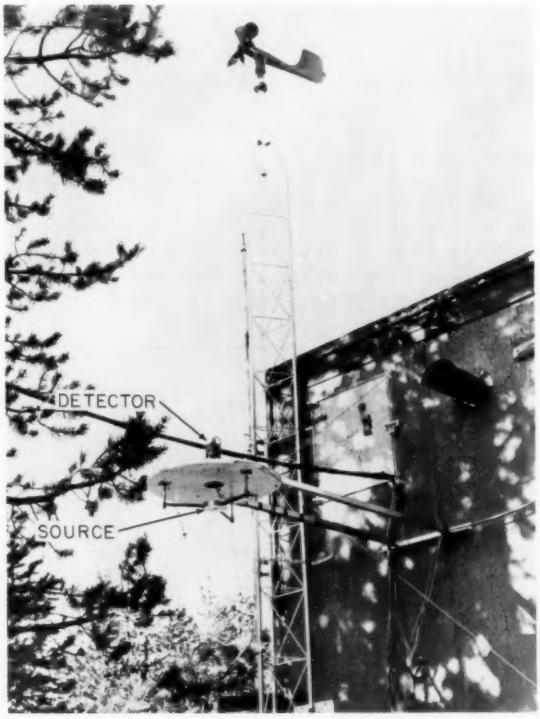

Fig. 2 Installation of radioisotopic rime-accumulation monitor.

*Forest Hydrology Laboratory, Pacific Northwest Forest and Range Experiment Station, Forest Service, U. S. Department of Agriculture, Wenatchee, Wash.

In Situ Elemental Analysis for Oceanographic Applications

By J. H. Reed* and J. W. Mandler*

Supplementary Keywords: *sea; industry, mining; activation analysis; source, neutron; activation in situ.*

The results of preliminary studies at the Illinois Institute of Technology Research Institute indicated that neutron activation analysis is a feasible technique for determining in situ the element composition of ocean-floor materials.

dunite saturated with water, and in dunite saturated with salt water (25 g of NaCl per kilogram of H_2O) were determined. In one series of tests, a radioactive neutron source (^{239}Pu–Be, 10^6 n/sec) was used with a 7-mm Ge(Li) detector and in the other, a 14-MeV pulsed-neutron source with a 3- by 3-in. NaI(Tl) detector. The detectors (surrounded by a ^{6}LiF thermal-neutron shield) were protected from the fast

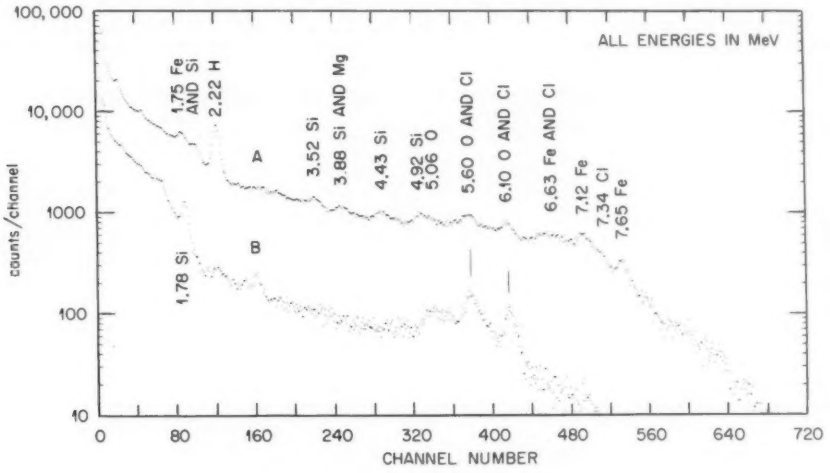

Fig. 1 Gamma-ray spectrum from dunite saturated with saline solution: 14-MeV pulsed-neutron source and NaI(Tl) detector. *A*, prompt gamma rays from neutron source recorded during a 200-μsec interval following the neutron pulse (capture plus cyclic activation). *B*, delayed gamma rays recorded in the 200-μsec interval preceding a neutron pulse (cyclic activation).

The technique was investigated on dunite, which is chiefly $(Mg,Fe)_2SiO_4$. A 43- by 24- by 12-in.-deep container filled with crushed dunite was surrounded by bags of crushed dunite to simulate a semi-infinite sample. The dunite was then neutron irradiated, and the gamma spectrums induced in the dry dunite, in neutrons and gamma rays produced in the neutron source by a cylindrical molybdenum shadow shield. The source–detector probes were placed on the sample and covered with a paraffin moderator, which thermalizes and reflects the fast neutrons. Since fewer neutrons are then captured by the water solution above the moderator, interferences from the oxygen, sodium, and chlorine in the water are decreased.

Spectrums taken with the pulsed-neutron source and the NaI(Tl) detector are shown in Fig. 1. The prompt-gamma-ray spectrum recorded with the Ge(Li)

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detector, using a radioisotopic neutron source, is shown in Fig. 2. The high resolution of the Ge(Li) detector largely obviates the advantage of pulsed operation, although the high-neutron machine source does have the advantages that it can be turned off and requires no shielding.

The technique is likewise suitable for analysis of manganese-containing nodules. (MG)

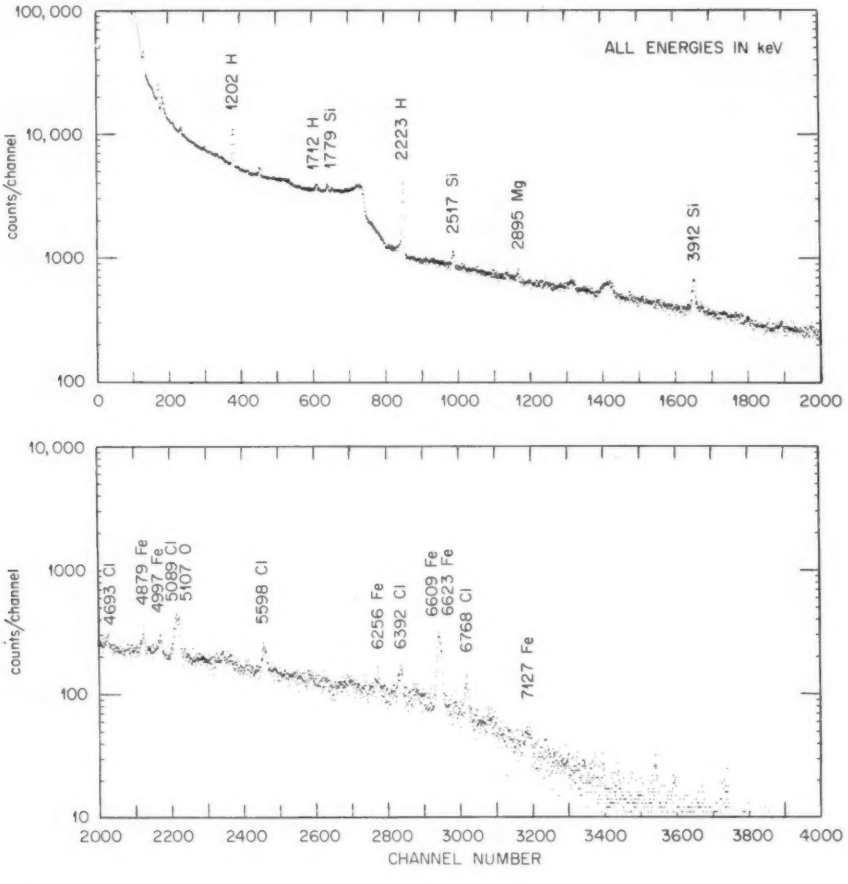

Fig. 2 Gamma-ray spectrum from dunite saturated with saline solution: isotopic neutron source and Ge(Li) detector.

A STUDY OF MINE DETECTION BY MEANS OF NEUTRON-INDUCED GAMMA RAYS

By F. R. Mynatt, R. G. Alsmiller, Jr., and L. R. Williams, USAEC Report ORNL-TM-3305, Oak Ridge National Laboratory, Jan. 11, 1971 (*Nuclear Technology*, November 1971).

The feasibility of locating land mines by detecting the photons induced in the mine by the neutrons from a fission source at the earth's surface was investigated by making neutron- and photon-transport calculations. The omnidirectional photon flux per unit energy when the mine is present was found to be significantly different from that when the mine is absent. However, the absolute photon intensity is so small that a rather large fission-source strength would be required to achieve an appreciable counting rate. (MG)

RADIOACTIVE ISOTOPES IN SCIENTIFIC RESEARCH

Report on Symposium held in Budapest, Nov. 12–19, 1970, *Atomtechnikai Tajekoztato* [Atomic Communications], **13**(1) (January 1970) (in Hungarian).

Abstracts of papers presented at the symposium and abbreviated versions of two papers from the plenary session are given. These two were "Use of Radioactive Isotopes for the Study of the Active Transport of Living Cells" by E. Broda (Institute of Physical Chemistry, Section of Radiation Chemistry of the University of Vienna), and "Effect of Radioactive Radiation on the Heterogeneous Equilibrium in the Solid-Solution System" by V. I. Spitsyn, E. A. Torchenkova, and I. N. Glazkova (Lomonosov State University, Moscow). The other 114 abstracts are of papers on physicochemical phenomenons, biochemical studies, tracer techniques in metallurgical and other industrial applications, and agricultural research using radioactive isotopes.

Some of the industrial studies included wear, metal surface temperatures (^{85}Kr), beneficiation of high-sulfur oils (^{35}S), silt movement, watermark labeling, and cellulose-manufacture processes. (François Kertesz, Information Division, ORNL)

GAMMA-RAY TECHNIQUES FOR MEASUREMENT OF SOIL DENSITY AND DENSITY PROFILE*

The U. S. Army Engineer Waterways Experiment Station, Vicksburg, Miss., has just completed a study to determine the feasibility of using measurements made with a multichannel gamma-ray spectrometer and a 10-mCi ^{60}Co source for accurately determining soil density and resolving the density profile of layers. Measurements were first made on aluminum and steel plates to establish a standard reference for the determinations.

Measurements were made on two samples of air-dried Yuma sand placed in a pit in thin layers. The density of the layers was controlled by the rate of flow of the sand and the distance the sand fell. The first pile was formed to a depth of 122 cm in a pit 51.82 by 3.54 m; the second was 125 cm deep. Measurements were made at depth intervals of 12.7 cm in each of six access holes located in the samples. The distance between the source and detector was 121.9 cm in all cases. Densities were compared with densities determined by nonnuclear means. The difference between the nuclear and nonnuclear methods was about 0.04 g/cm^3 at a density of 1.5 g/cm^3.

Results of the study indicate that density can be measured accurately by this method provided (1) the thickness through which the measurements are made is accurately measured and (2) the source strength and detector are suitable for the distance over which the density is measured. The combination of source and detector that was used permitted defining soil-density profiles.

As a result of this study, the nuclear method was recommended for routine measurements of subsurface soil density. Further feasibility studies on the method were not recommended, since it is based on a well-understood physical phenomenon. (RHL)

*Extract from A. N. Williamson, Jr., Gamma-Ray Techniques for Nondestructive Measurements of Soil Density and Density Profile, Technical Report M-70-14, U. S. Army Engineer Waterways Experiment Station, November 1970.

Section III

Isotopes and Radiation Technology

Food Irradiation

Status of Approval of Irradiated Food Products for Human Consumption

Supplementary Keywords: *industry, food; wholesomeness; regulation; accelerator;* ^{60}Co; ^{137}Cs.

An International Atomic Energy Agency survey [*International Atomic Energy Agency Bulletin*, **13**(1): 24-25 (1971)] shows that the countries listed below have approved various irradiated food products for human consumption:

Country	Product	Purpose of irradiation	Radiation source	Permissible dose, rads	Approval date
Canada	Potatoes	Sprout inhibition	^{60}Co	10 max 15 max	Nov. 9, 1960 June 14, 1963
	Onions	Sprout inhibition	^{60}Co	15 max	Mar. 25, 1965
	Wheat, wheat products	Insect disinfestation	^{60}Co	75 max	Feb. 28, 1969
Denmark*	White potatoes	Sprout inhibition	Electron accelerator	15 max	Jan. 27, 1970
Israel	Potatoes	Sprout inhibition	^{60}Co	15 max	July 5, 1967
	Onions	Sprout inhibition	^{60}Co	10 max	July 25, 1968
Netherlands	Asparagus (experimental batches)	Radurization	^{60}Co	200 max	May 7, 1969
	Cacaobeans (experimental batches)	Insect disinfestation	^{60}Co, 4-MeV electrons	70 max	May 7, 1969
	Strawberries (experimental batches)	Radurization	^{60}Co, 4-MeV electrons	250 max	May 7, 1969

*Information later than reference cited.

(Tabulation continues on the next page.)

Country	Product	Purpose of irradiation	Radiation source	Permissible dose, rads	Approval date
Netherlands (Continued)	Mushrooms	Radurization	^{60}Co, 4-MeV electrons	250 max	Oct. 23, 1969
	Potatoes	Sprout inhibition	^{60}Co, 4-MeV electrons	15 max	Mar. 23, 1970
Spain	Potatoes	Sprout inhibition	^{60}Co	5 to 15	Nov. 4, 1969
USA	Wheat, wheat products	Insect disinfestation	^{60}Co ^{137}Cs 5-MeV electrons	20 to 50 20 to 50 20 to 50	Aug. 21, 1963 Oct. 2, 1964 Feb. 26, 1966
	White potatoes	Sprout inhibition	^{60}Co ^{137}Cs ^{60}Co, ^{137}Cs	5 to 10 5 to 10 5 to 15	June 30, 1964 Oct. 2, 1964 Nov. 1, 1965
USSR	Potatoes	Sprout inhibition	^{60}Co	10	Mar. 14, 1958
	Grain	Insect disinfestation	^{60}Co	30	1959
	Dried fruits	Insect disinfestation	^{60}Co	100	Feb. 15, 1966
	Dry food concentrates	Insect disinfestation	^{60}Co	70	June 6, 1966
	Fresh fruits and vegetables (experimental batches)	Radurization	^{60}Co	200 to 400	July 11, 1964
	Semiprepared raw beef, pork, and rabbit products in plastic bags (experimental batches)	Radurization	^{60}Co	600 to 800	July 11, 1964
	Eviscerated poultry in plastic bags (experimental batches)	Radurization	^{60}Co	600	July 4, 1966
	Culinary prepared meat products (fried meat, entrecôte) in plastic bags (experimental batches)	Radurization	^{60}Co	800	Feb. 1, 1967
	Onions (experimental batches)	Sprout inhibition	^{60}Co	6	Feb. 25, 1967

(MG)

Wholesomeness of Irradiated Food, Especially Potatoes, Wheat, and Onions*

Summarized by F. E. McKinney

Supplementary Keywords: *food preservation; grain; potato and onion; industry, food.*

Abstract: *A recent World Health Organization report of an expert committee on wholesomeness of some irradiated foods is summarized. The committee recommended that irradiated wheat and potatoes be given temporary acceptances because no positive evidence of harmfulness has been found; however, they recommended further feeding studies to determine effects on reproduction and nutrient content for wheat and on reproduction and carcinogenicity for potatoes. The committee decided that insufficient data are available at this time on which to base a decision for irradiated onions.*

An expert committee on wholesomeness of irradiated food met in Geneva from Apr. 8–12, 1969. The committee was made up of members invited by the World Health Organization (WHO), the Food and Agricultural Organization of the United Nations (FAO), and the International Atomic Energy Agency (IAEA). The members invited by WHO evaluated the data on wholesomeness of wheat irradiated for disinfestation and of potatoes and onions irradiated to control sprouting. The members invited by FAO and IAEA discussed technical aspects and provided information required by the WHO members. The decision of the committee was based entirely on toxicological and nutritional data; technological and economic questions were not considered.

DATA EXTRAPOLATION

The committee had to evaluate data obtained under various sets of conditions, and it had to take into account that the food as eaten by man may be different in form from that eaten by test animals. The relationship between radiation dose and the chemical effects of radiation on food has not been clearly established. Evidence suggests that a plateau effect exists for the concentration of radiolytic products and that a threshold may exist below which change cannot be detected. The committee decided that toxicological evidence from animals fed food irradiated at doses much higher than those to be used in practice might not be appropriate, although data from high-dose experiments may indicate effects to be looked for in experiments at more practical doses.

There are quantitative differences in composition between varieties of a crop and between crops of the same variety grown in different locations; the basic composition is likely to be qualitatively similar. The committee decided that for irradiated wheat and potatoes the evaluations should have general applicability, regardless of variety or area where grown.

Differences in physical form of a food can influence the radiation-induced chemical changes, but the committee felt that these differences would be quantitative rather than qualitative. Data on both wheat grain and wheat flour were taken to be applicable to both forms of wheat and wheat products.

MUTAGENICITY AND CYTOTOXICITY

Since the report of an FAO/WHO/IAEA expert committee[1] in 1964, published information has shown that cytotoxic and mutagenic substances may be formed in irradiated food. Both gene mutations and chromosome aberrations may be induced. Tests are being developed to assess the mutagenic properties of the induced substances. In view of the scarcity of data and the uncertainty of its extrapolation to man, the committee discounted the available mutagenic data in reaching its decision on the wholesomeness of specific irradiated foods. Mutagenic substances might also result from other food processes and certain accepted food additives and other materials.

INVESTIGATIONS REQUIRED FOR ASSESSMENT OF WHOLESOMENESS

The committee endorsed the opinion expressed in the 1964 meeting[1] that the principles of food-additive testing are inapplicable to irradiated foods insofar as

*A summary of "Wholesomeness of Irradiated Food with Special Reference to Wheat, Potatoes, and Onions," Report of a Joint FAO/IAEA/WHO Expert Committee, Geneva, Apr. 8–12, 1969, World Health Organization Technical Report No. 451, 1970.

physical and chemical identification are concerned. The methods recommended in the 1964 report are still valid, although certain modifications and elaborations were suggested. Collaboration between the appropriate government authorities and investigators is essential.

Experimental Design

When long-term investigations are conducted, a positive control group should be included and fed a known carcinogen to check the response of the species used. Consideration should be given to the use of several control groups; for instance, an additional group fed the same food treated by alternative accepted methods, such as thermal processing, might yield useful comparative information. The committee questioned the suitability of chickens for use as test animals in long-term tests because insufficient toxicological experience is available with this species. In carcinogenicity studies, the use of rats and mice is preferable. Recent experience with the miniature pig is not encouraging, and there is an urgent need for the discovery of other nonrodent species suitable for long-term studies. Feeding studies with large animals, such as dogs and pigs, have frequently used too few animals. The minimum number of large animals should be eight.

Many irradiated food studies have produced poor results because insufficient attention has been given to the nutrient and caloric value of the diet. The diets should be isocaloric, be of adequate caloric density for the test species, and have a nutrient level equal to the normal requirements of the test species. Before the diet is fed, it should be subjected to the same conditions and procedures envisaged in practice. Any additional process applied to the control diet must also be applied to the irradiated diet. The radiation source, the energy, the radiation dose, and the radiation dose rate should be specified in the report, along with full details of the environmental conditions during and following irradiation.

Experience has shown that dogs are generally unsuited for routine studies of reproductive function. Two species are required; mice and rats are acceptable, and Japanese quail are sometimes useful.[2] The teratological examinations recommended by an expert committee[3] in 1966 can be undertaken by breeding third litters in a multigeneration test and terminating pregnancy a few hours or a full day before expected delivery so that all surviving fetuses can be examined and the resorption sites and number of dead fetuses counted.

EVALUATION OF IRRADIATED FOODS

The committee examined the extent and quality of the biological evidence and other available data in making their decisions. The usual consumption of the specific food in different countries and its role in nutrition were also considered. Three categories of acceptance were established: (1) *unconditional acceptance*, which is granted where adequate data are available for full evaluation; (2) *conditional acceptance*, which is granted where data are not sufficient for full evaluation (further investigations are usually specified); and (3) *temporary acceptance*, which is granted where there are insufficient data and where additional information is required within a stated period of time (if the data do not become available, the acceptance will be withdrawn). Sometimes *no decision* can be made because the available evidence is inadequate or unsuitable; a *no decision* does not imply that there is evidence of harmfulness. The committee will recommend *nonacceptance* if available evidence demonstrates a potential health hazard.

The specific conditions evaluated were: (1) wheat or ground wheat products irradiated to a maximum dose of 75 krads with gamma radiation from ^{60}Co or ^{137}Cs or with electrons of less than 10 MeV; (2) white potatoes irradiated with gamma radiation from ^{60}Co or ^{137}Cs to a dose of 15 krads; and (3) onions irradiated with gamma radiation from ^{60}Co or ^{137}Cs to a dose of 15 krads. The doses specified are maximum doses chosen after considering the overdosing that might occur in practice under plant conditions. The actual dose used in practice will be as small as possible, for economic reasons, and will be set by the plant management. The use of electrons was considered only in some instances, and the use of X rays not at all, because there is insufficient experience in their technological application.

IRRADIATED WHEAT*

The purpose of irradiating wheat is to control infestation by insects in stored grain and in ground wheat products.† The dose required depends on the insect species to be controlled and on the permissible

*As in other WHO reports on wholesomeness of irradiated food, separate monographs were prepared for the specific foods and attached to the report as appendices. They are inserted into this summary to provide continuity.

†The conclusions in this section are supported in the summarized WHO report by a bibliography of 46 references.—Editor's Note.

time interval between irradiation and death of the insects. For most species, a 10- to 25-krad dose is sufficient; mites and certain moths require higher doses. For evaluation, a maximum of 75 krads was assumed.

Nutritional Effects

The protein content, measured as total nitrogen, is unaffected by radiation. Amino acid content is also unaffected by practical doses, although small losses in leucine, isoleucine, and methionine occur at 5 Mrads. The protein nutritive value of whole wheat is unaffected by a 200-krad dose, but statistically significant losses occur with doses of 1 Mrad or higher. The nutritive value of gluten prepared from wheat and then irradiated is unaffected at 20 krads but shows a 5% loss at 200 krads, principally because of decreased availability of methionine. Tests in rats have shown that protein-efficiency ratio, nitrogen excretion, and digestibility are unaffected at 20 krads. Digestibility and biological value of wheat gluten irradiated to 2.8 Mrads were similar to those of untreated gluten. Irradiation of wheat bran with 0.4 or 5.0 Mrads increased net protein utilization in chickens. The increase in maltose values of irradiated wheat at doses greater than 0.5 Mrad is attributed to depolymerization of polysaccharides.

The evidence on B vitamins is plentiful but sometimes conflicting. No loss of thiamine was detected in whole wheat after doses of 20 or 200 krads or in flour after 70 krads. Other researchers noted small losses in flour after doses of 70 krads or more. There is some evidence that thiamine loss is dose dependent up to 500 krads and independent of moisture content. Other researchers have noted considerable losses of thiamine. No loss of riboflavin in wheat flour was noted up to 74 krads or in wheat grain up to 200 krads. Even after 5 Mrads, no loss was evident in a grain diet for chickens. No loss of nicotinic acid in grain at 20 krads or in flour at 37.2 krads was detected, but small losses were reported in flour at 50 and 74.4 krads. A 12% loss was reported in grain after a dose of 200 krads. No significant loss of total vitamin B_6 occurred in grain after a 200-krad dose, although a chick diet lost 20 to 30% after a 5-Mrad dose. No loss of biotin occurred at 20 krads, but a loss of 10% was found after a dose of 200 krads. Losses of pantothenic acid were detectable after 20 and 200 krads, but they were too small to be of nutritional concern.

Vitamin E is very susceptible to radiation in some foods, but only a small loss has been observed in whole-meal flour prepared from irradiated wheat at doses up to 200 krads. Rats whose only source of vitamin E was wheat irradiated at 10 krads showed no deficiency symptoms through four generations. A specific-pathogen-free breeding colony was maintained for 4 years on an unsupplemented laboratory chow sterilized with 2.5 Mrads; the breeding performance, usually taken as an indicator of vitamin E availability, was excellent.

Irradiation of wheat bran caused an increase in net phosphorus utilization and absolute retention of phosphorus by chickens. Phosphorus in the polar fraction increased in lipids of flours milled from wheat irradiated at high doses. No loss of ascorbic acid was noted in flour irradiated at 80 krads.

Feeding Studies

Mice. No differences in number born or in development of young were observed in a five-generation test with wheat irradiated to 27.9 krads. In a 2-year study of two strains of mice, all mice showed a degree of breeding failure but there was no effect on body weight or survival. In one strain, 28 mice developed lymphosarcomas—including 11 of the control group. Reproductive capacity was unimpaired in a multigeneration test of a 75% wheat diet irradiated at 20 or 200 krads. One long-term study of wheat irradiated to 5 Mrads did show cytogenetic abnormalities in developing spermatogonia, but this dose is considerably higher than would be used in practice. There was no effect on fecundity, although losses after birth were higher in the group fed irradiated flour.

Rats. Wheat irradiated to 20 or 200 krads in a mixed diet produced no effects on body weight or feed efficiency. Four generations were fed wheat irradiated to 10 krads and stored under adverse conditions; male fertility was reduced and infant mortality increased, but the effect was greater in the control animals than in the group on the irradiated diet. In a massive experiment with four generations fed a diet of 75% wheat irradiated to 20 or 200 krads, no effects were noted on fertility, number of young, sex ratio, survival after birth, or weight after birth. Comprehensive analysis of 1238 autopsies gave no evidence that wheat is tumorigenic. In another experiment, over a life-span plus five generations, a diet of 75% wheat irradiated at 20 or 200 krads showed no effect on growth, feed efficiency, intestinal flora, or reproduction. In one experiment, flour was irradiated to 37 or 74 krads, stored for 3 months, and then cooked before feeding. No effects were evident in the test animals after 2 years, and independent examination of histological

material confirmed that there was no increase in tumors. Other experiments showed no effect on blood characteristics and blood enzyme systems. Experiments with wheat irradiated with spent fuel elements to doses of 2.8 and 5.6 Mrads showed that rats on irradiated diet actually gained more weight than control animals; this was attributed to increased availability of nutrients caused by degradation of cellulose.

Chickens and Other Animals. Chickens fed cereal grains irradiated to 19.5 krads showed no differences in body weight, egg production, egg weight, or egg quality. Germ-free chicks fed a 30% ground wheat mash sterilized with 5 Mrads grew better than ordinary control chicks, but ordinary chicks on the irradiated diet did no better than the controls. Hens on a radiation-sterilized mash did show a slightly lower growth rate caused by a slight vitamin deficiency in the diet. Pigeons fed grain irradiated to 100 krads showed no differences in reproduction function. Both control dogs and dogs fed irradiated flour became obese on a diet of 35% flour.

Man. Two controlled series of human feeding studies have been reported. Foods of all kinds irradiated with spent fuel elements were fed in 15-day periods to young male conscientious objectors. In the first study, diets of 35, 60, 80, and 100% irradiated food were fed in successive periods, alternating with similar periods in which unirradiated foods were fed; each subject thus served as his own control. This first study included bread and other cereal products irradiated to 2.8 Mrads. The second study included flour irradiated at 75 krads and incorporated as bread and confectionery items in a diet that was about 80% irradiated. The studies failed to demonstrate any deleterious effects on general health or damage to organs studied. There was no evidence of clinical abnormality that could be related to the ingestion of irradiated food.

Recommendations on Irradiated Wheat

The disturbing effects noted in mice fed wheat irradiated with 5 Mrads are the basis for the requirement that certain further work be undertaken to confirm that similar effects on reproduction are not demonstrable at dose levels of practical importance. The evidence concerning nutritive adequacy of irradiated wheat and ground wheat products does not give cause for concern. The committee noted a lack of information on vitamin content of bread prepared from irradiated wheat. The recommendation is, therefore, that wheat and wheat products be given a *temporary acceptance* with further studies to be completed within 5 years. The additional studies must demonstrate the absence of adverse effects on reproduction and must assess the nutrient content of bread prepared from flour stored for periods up to 2 years after irradiation before use. These additional studies must be completed before Apr. 30, 1974.

IRRADIATED POTATOES*

Potatoes are irradiated to inhibit sprouting during storage. The dose varies slightly with variety; some varieties need only 7 to 10 krads. The maximum dose chosen for this study was 15 krads.

Nutritional Effects

A slight loss of vitamin C occurs during and immediately after irradiation, with the loss being dose dependent. At 10 krads, some researchers have reported 15% loss, while others have reported no loss in ascorbic acid. The main cause of loss of ascorbic acid in potatoes is storage; after 4 months, there is usually little difference between irradiated and nonirradiated potatoes.

Irradiation does cause an immediate increase in reducing sugars and sucrose. If potatoes are then stored at a low temperature (4°C), after 4.5 months the sugar content will be comparable to that of nonirradiated potatoes. If potatoes are stored at high temperature, the sugar content will remain high, with a correspondingly low starch content. The formation of deoxyglucose from starch has been suggested as a reaction that could be used to detect whether or not potatoes have been irradiated.

Doses up to 20 krads do not significantly alter amino acid composition of potato proteins. Like sugars, proteins show an immediate effect after irradiation, but subsequent storage reduces these differences between control and irradiated potatoes.

Feeding Studies

Rats. After a dose of 9 krads, given at an exceptionally low dose rate, irradiated potatoes produced no changes in growth, blood, or reproduction. A diet of 53% raw potatoes was unsuitable and caused physiological disturbances in control groups and

*The statements in this section are supported in the original WHO report by a bibliography of 22 references.—Editor's Note.

groups fed 200-krad irradiated potatoes, but reproduction was normal, and testes of males showed no differences in activity of germinal epithelium. A diet of 72% dried potatoes irradiated to 10 krads with electrons gave no differences in weight gain or protein efficiency after 1 year. A short-term study showed that 8 krads increased digestibility of dry matter and nitrogen-free extracts slightly, but there was a decrease in total digestive nutrients and net energy.

In a 14-month study, potatoes irradiated to 46.5 krads produced no effects on mortality, growth, blood characteristics, urine, or reproduction. Some variations in female fertility and pup weights occurred in a 2-year study covering four generations; potatoes were irradiated at 13.5 to 20 krads and at 27 to 40 krads. There was no effect on litter size or survival until weaning, and growth and feed efficiency were normal. Coronary arteriosclerosis, focal myocarditis, and bronchiectasis were more frequent in rats on the irradiated diet. Frequency of tumors was similar in control and test groups. An independent analysis came to the same conclusion, although different figures resulted. Another four-generation study, evaluated independently because of inconsistencies and errors in calculation, showed a slightly higher congestion in spleen and occasional minor lesions in rats on the higher dose diet.

Dogs. No effects of any kind were found in dogs fed potatoes irradiated to 46.5 krads. Dogs fed for 101 weeks on a 35% (dry weight) potato diet, with doses of 7 and 14 krads, did not achieve normal body weights, but control and test-group weights were comparable on this suboptimal diet.

Pigs. Pigs fed irradiated potatoes for 13 weeks showed no differences from controls; bacon was similar from both control and test groups. Adult pigs fed 4 months on potatoes irradiated to 15 krads showed no differences from controls. Piglets raised to maturity on a diet of irradiated potatoes showed no effects on fertility, number of offspring, or weight of offspring. A large feeding study failed to confirm any consistent differences in a large number of blood characteristics.

Recommendations on Irradiated Potatoes

Few long-term feeding studies were considered satisfactory. However, the committee did not find any evidence that irradiated potatoes are harmful. It recommended *temporary acceptance* of irradiated potatoes, with further feeding studies required by Apr. 30, 1974, to assess in rats and mice any effect on reproductive function. Carcinogenicity must be assessed in a second species, e.g., mice.

IRRADIATED ONIONS*

Onions are irradiated to inhibit sprouting. The usual dose is between 8 and 10 krads.

Nutritional Effects

Doses up to 12 krads had little effect on ascorbic acid content of onions at intervals up to 7.5 months after irradiation, and there was no conversion of reduced ascorbic acid to the dehydro form. Doses of 14.9 krads caused no consistent effects on total sugar content and reducing sugars during storage at 10°C, although some workers have reported an accumulation of nonreducing sugars in irradiated bulbs.

Feeding Studies

Rats. A diet of 3% irradiated onion (dry weight, equivalent to 25% wet weight) was fed for 12 weeks; growth was normal, but erythrocyte counts and hemoglobin levels were lower in rats fed the onions dosed with 20.3 krads. An experiment with onions incorporated at 10 and 25% wet-weight levels after doses of 8 and 100 krads showed no effects on growth, reproduction, or blood picture after 1 year and three generations. An experiment with onions incorporated at a 35% dry-weight level after a dose of 25 krads showed that both test and control groups had severely affected spleens and livers at this extremely high level of onion.

Dogs. Urinalysis of beagles fed for 90 days on 10% dried onion irradiated to 25 krads showed no effect, but hematological examinations were inconclusive because of anemia in both control and test animals. All dogs had increased spleen weights. Another test with onion incorporated at a 25% wet-weight level showed that all dogs on onion, irradiated to 20 krads or not, tended to become anemic, had liver and spleen damage, and showed varying degrees of ovarian or testicular degeneration.

Pigs. Autopsies of pigs fed for 24 weeks on onions at a 25% wet-weight level, and after a dose of 20.3 krads, showed osteodystrophy due to a calcium and phosphorus imbalance in both control and diet groups; weight gains and food intake were below normal. Hematocrit values were higher in animals on irradiated diets.

*The statements in this section are supported in the summarized WHO report by a bibliography of seven references.—Editor's Note.

Recommendations on Irradiated Onions

Because onions contain constituents that cause hemolysis and anemia in many species of animal, feeding diets containing large amounts of onions is difficult. The appearance of toxic effects does not mean that irradiation has caused some change in the onions. The committee recommended that a search be made for a species that can tolerate high content of onions in the diet or for an alternative test method. If levels of dietary intake are necessarily low, then larger numbers of animals than usual may have to be used.

Data available on wholesomeness of irradiated onions are not sufficient to allow an evaluation. For instance, there are no long-term feeding studies on onions. The committee handed down a *no decision* ruling on irradiated onions.

FUTURE RESEARCH

Man is continually exposed to potentially harmful materials, and food is only one factor in the total chemical contamination of his environment. The potential toxicity of irradiated food should not be viewed in isolation. Irradiation could replace certain pesticides that are toxic; this is a strong reason why further exploration in the use of radiation to preserve food should be encouraged and supported.

Animal feeding studies should be continued on individual irradiated foods. Irradiation should be performed under practical conditions and on food in forms closely comparable to those consumed by man. A degree of extrapolation will be necessary because of variations in irradiation facilities and of differences in preparation methods of certain foods. Generally applicable data are accumulating from specific studies, but studies designed specifically for this purpose should still be carried out. Studies on foods that represent broad classes of food should be made, and consideration should be given to both composite diets and extract or concentrate diets. Radiation doses should be within the range of practice. Each study should attempt to identify and estimate the products resulting from irradiation of the food. Details concerning the type of radiation, total dose, dose rate, and dose distribution should be provided along with information on the conditions of irradiation (temperature, atmosphere). Research into the effects of radiation on nutritional value should be continued, especially on irradiated foods as consumed in practice. Further research into the microbiological aspects is also recommended.

New technical procedures need to be developed for testing irradiated foods. Modifications in immunological responses and alterations in behavior patterns after consumption of irradiated food are possible sensitive indexes of toxicity. Species other than rodents need to be developed for carcinogenicity studies, and further work on methods of testing for mutagenicity is required.

National legislation governing production and sale of irradiated food must be enforceable. Control by means of licensing and label declarations would be strengthened if there were some means to test food to determine if it had been irradiated. Research into the development of such methods should be continued.

RECOMMENDATIONS

Further meetings of WHO, in consultation with IAEA and FAO, should be convened on these specific foods and on revision and clarification of existing documents for the technical basis for legislation. Responsible international agencies should collect data pertaining to wholesomeness, provide consultation on wholesomeness studies, and encourage research on new methods for testing wholesomeness. The committee also recommended that WHO convene a meeting of experts on the assessment of mutagens; such a meeting could guide future research on possible mutagenic hazards due to natural components of food and the consumption of food containing additives as well as food subjected to various processing methods, such as irradiation.

REFERENCES

References marked with an asterisk have not been verified by the editors; for further explanation, see the Foreword.

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Effects of Low-Level Irradiation on the Preservation of Fruit: A 7-Year Summary*

By R. A. Dennison and E. M. Ahmed*

(Summarized by F. E. McKinney)

Supplementary Keywords: *food preservation; industry, food.*

Abstract: *Work was conducted under a USAEC Division of Isotopes Development contract from May 1963 through February 1970 to determine the effect of low-level irradiation upon certain food items prominent in the Florida agricultural economic system, with specific reference to citrus. Work was conducted to determine the effects of irradiation on the decay, peel injury, weight loss, firmness, color, flavor, composition, yield and quality of juice, respiration, and behavior during shipping of citrus fruits. Summary results are also presented on the effects of low-dose irradiation on peaches, mangoes, and tomatoes. Microbiological studies examined the effects of low-dose irradiation of oranges and lemons inoculated with stem-end decay fungus and of peaches inoculated with brown-rot fungus. This article is a summary of USAEC Report ORO-680.*

The Department of Food Science of the University of Florida, Gainesville, held a contract from the Division of Isotopes Development, USAEC, for the period May 1963 through February 1970 to investigate the effects of radiation on food items important to the Florida agricultural system, with specific emphasis on citrus fruits. This article presents the main findings that resulted from this period of research. The work was carried out using a pool-type irradiator, originally containing ^{60}Co; in 1968 the cobalt was replaced by ^{137}Cs. Because of the final summary nature of the article, a bibliography of articles and reports based on data accumulated under this contract is included rather than a list of specifically called-out references.

CITRUS FRUITS

Stem-end rots and green and blue molds cause the greatest postharvest decay losses to citrus fruits. No single radiation dose can be given to protect these fruits against spoilage, and the dose required for retarding established infections is higher than that for retarding incipient infections. The peel of citrus fruits is very sensitive to radiation damage, especially at the doses required for decay control. The extent and severity of peel injury increases with the duration of storage and with higher storage temperature.

Decay Control

There was considerable variation in the control of decay by irradiation; in some tests the amount of spoilage was as great or greater in irradiated lots as in control lots. Some of the factors that were associated with the variation in decay control were time of harvest, maturity of the fruit, whether or not the fruit had been waxed, and variety of fruit. Dipping the fruit in hot water (127°F for 5 min) prior to irradiation reduced the incidence of decay in most tests. Typical variations in decay control found with Temple fruit harvested at different dates, field run or processed (washed and waxed); and dipped or not are given in Table 1.

Peel Injury

Peel injury starts with swelling of the oil glands and results in pitting of the peel, mainly around the stem end of the fruits. Irradiation tends to increase peel injury.

An effort was made to find methods for reducing the extent of peel injury caused by irradiation. Storing fruit at 35°F, rather than 50°F or higher, prevented the fruit from showing large amounts of peel injury. However, when fruit stored at 35°F was moved to temperatures typical of supermarkets (78°F), the injury on the surface of the peel increased. When fruit was dipped in hot water (127°F) for 5 min, cooled in tap water, and then irradiated, the peel injury was reduced.

Peel injury ratings for different treatments, using Navel oranges, are given in Table 2. In some tests, the severity of peel injury was greater than shown in this table.

*Department of Food Science, University of Florida, Gainesville, Fla. Original title: Effects of Low Level Irradiation Upon the Preservation of Food Products, Final Summary Report for the Period May 1963–March 1970.

Table 1 Variations in Decay of Temple Fruit After Irradiation

Harvest and storage	Dose, krads	Decay, %			
		Field run, not dipped	Processed, not dipped	Field run, dipped*	Processed, dipped
1/29/69;	0	5.0	15.0	1.7	3.3
12 days at	100	11.7	21.7	3.3	1.7
65°F	150	8.3	23.3	0.0	5.0
2/13/69;	0	21.7	40.0	20.0	55.0
13 days at	100	8.3	23.3	16.7	21.7
65°F	150	6.7	2.17	10.0	5.0

*Dip treatment was hot water at 127°F for 5 min.

Table 2 Peel Injury Ratings for Various Treatments of Navel Oranges

Storage regime	Dose, krads	Injury rating*	
		Not dipped	Dipped†
21 days	0	10.0	10.0
at 35°F	50	10.0	10.0
	100	10.0	10.3
	150	10.3	11.0
21 days	0	10.0	10.0
at 65°F	50	16.8	13.3
	100	16.0	16.4
	150	17.5	14.9
21 days	0	10.0	10.0
at 35°F plus	50	19.3	13.0
7 days at	100	21.4	14.5
78°F	150	24.7	13.9

*Injury scale: 10 = none, 20 = slight, 30 = moderate, 40 = severe.
†Hot water at 127°F for 5 min.

Weight Loss

There was no consistent trend in weight loss with the treatments used, except for the progressive increase with duration of storage. In most tests there was no significant difference in the weight lost by the irradiated and the nonirradiated fruit.

Firmness

Whole fruit and fruit sections of irradiated Temple fruit were softer than nonirradiated fruit. The greatest decrease in firmness was noted immediately after irradiation, but firmness differences between irradiated and nonirradiated fruit were smaller after storage.

Color

Irradiation accelerated the rate of external color change from green to yellow of harvested Bearss lemons initially, but the increase did not continue throughout the holding period. The nonirradiated and irradiated lemons reached the full yellow color at approximately the same time.

Flavor

Flavor scores for juice expressed from irradiated fruit usually decreased (Table 3). The greatest flavor differences were noted immediately following irradiation, but as the storage period increased, differences were fewer. Doses up to 200 krads in most tests did not cause extreme changes in flavor; above this dose, juice expressed from irradiated fruit was almost always unacceptable.

Table 3 Flavor Scores of Juice from Irradiated Temple Fruit

Storage at 35°F, days	Ratings*		
	Unirradiated	100 krads	200 krads
0	2.9	2.8	2.1
7	2.8	2.8	2.2
14	2.6	2.6	2.3
21	2.7	2.6	2.3

*Scores: 1 = poor, 2 = fair, 3 = good.

Composition

Irradiated fruit usually shows some loss of reduced ascorbic acid in the juice, but this loss seldom reaches as much as 10% of the total content. Typical changes

in reduced ascorbic acid content with irradiation and subsequent storage of the fruit are shown in Table 4. No consistent changes were found in amount of reducing sugars, sucrose, and total sugars.

Table 4 Changes in Reduced Ascorbic Acid Content of Tangerines

Dose, krads	Storage, days	Reduced ascorbic acid (T = 35°), mg/100 ml	Reduced ascorbic acid (T = 50°), mg/100 ml
0	0	31.19	31.19
	10	28.13	27.65
	20	27.71	24.71
40	0	32.48	32.48
	10	27.68	26.93
	20	25.51	24.78
80	0	29.19	29.19
	10	26.26	25.88
	20	23.85	23.10
160	0	29.42	29.42
	10	25.23	24.07
	20	23.08	20.20

Pectic fractions in the peel, membrane, and juice sacs of Valencia oranges and Duncan grapefruit changed with irradiation. Water- and oxalate-soluble pectins increased, but protopectin decreased. The least change in the fractions was found in the juice sacs. As dose increased, the amount of change in the fractions increased. Acceleration of pectic changes caused more rapid softening of the fruit than that which normally occurs.

The extent of depolymerization of the pectin molecule was determined by jelly-grade measurements. The greatest decrease in the jelly grades of the pectins was found in the peel and membrane when the Brix/acid ratio was lowest. The less mature the fruit, the lower the Brix/acid ratio.

Juice Yield and Quality

No differences in juice yield from Valencia oranges were caused by irradiation treatment. In some tests with Duncan grapefruit, there was a greater yield of juice from irradiated fruit, usually with more mature fruit.

Higher viscosity was exhibited by the juice expressed from the irradiated fruit as compared with the nonirradiated fruit (Table 5). Irradiation did not affect titratable acidity or solids content of the juice.

Table 5 Changes in Viscosity of Juice from Irradiated Duncan Grapefruit

Storage, days	Viscosity, cp			
	Unirradiated	100 krads	200 krads	300 krads
0	7.5	8.6	9.5	10.5
7	7.7	8.2	10.1	10.2
14	8.4	8.6	9.6	10.7
21	9.7	10.8	10.4	12.0

Respiration

In general, citrus fruits show an immediate increase in O_2 uptake and production of CO_2 when irradiated. The higher the irradiation dose, usually the higher the respiratory activity.

Shipping Studies

The responses of Valencia and Pineapple oranges and Temple fruit to simulated commercial shipping were evaluated in conjunction with the Department of Horticulture at the University of Maryland. Field-run fruits were washed, sized, dipped in hot water (5 min at 127°F), cooled in tap water, placed in 5-lb plastic bags, and irradiated. Half the bags were transported to College Park, Md.; the remainder were retained in the Department of Food Science at the University of Florida. Peel injury, weight loss, soluble solids, titratable acidity, *p*H, and reduced ascorbic acid were evaluated periodically.

Irradiated fruit had higher indexes of peel injury than nonirradiated fruit. In general, there was no indication that peel injury was increased by shipping. No general trends were observed in weight losses or in changes in solids content that could be attributed to irradiation or to stresses during shipment. Some irradiated fruits had lower acidities after prolonged storage, but shipping did not appear to influence the acidity. Losses in ascorbic acid due to irradiation ranged from 5 to 15%; shipping did not seem to affect ascorbic acid content.

PEACHES

Spoilage losses of peaches are caused mostly by infection from *Monolinia fructicola* (Wint.) Honey, commonly referred to as brown rot. Retardation of this infection would improve the marketing of fresh peaches. Irradiation has shown some promise as one method for control.

Decay Control

Methods investigated for decreasing spoilage were the use of radiation alone and of combination treatments of radiation and hot water. Both methods significantly reduced the number of decayed fruit, but the combination treatments gave the better control. Table 6 shows that a 75-krad dose did not control brown rot, but with a 7-min 120°F hot-water treatment, it did aid in lowering the amount of spoilage. When the dose was increased to 150 krads, a significant decrease in spoilage was obtained without the hot-water treatment.

Table 6 Decay Control in Firm Ripe Loring Peaches Stored 10 Days After Treatment at 35°F Plus 4 Days at 68°F

Dose, krads	Decay, %	
	No dip	Dip*
0	53.3	4.4
75	60.3	2.2
150	2.2	0.0
225	8.9	0.0

*Hot water at 120°F for 7 min.

Firmness

The stage of maturity and the variety of the peaches influence the firmness changes that result from irradiation and from combination treatments. In most tests, peaches are softer after irradiation. If they are irradiated at the mature green stage, there is a sharp drop immediately in firmness. At riper stages, only minor differences in firmness between irradiated and nonirradiated peaches occur. If they are irradiated at the firm ripe stage, the decrease in firmness is slight. A greater decrease in firmness results with the use of hot-water dips.

Skin Color

When peaches were irradiated at the mature green stage, visual observations during storage showed that the red coloration became more intense than in nonirradiated fruit. Objective color measurements verified the visual observations and showed that the increase in red color was accompanied by a decrease in green color. Pigment extractions showed an increase in anthocyanin (Table 7). Amount of skin used and volumes of crude extract were held constant so that absorbancy values would be on a comparable basis. When peaches were irradiated at the firm ripe stage, differences in pigmentation did not occur.

Biochemical Changes

Some varieties of peaches, but not all, showed slight losses in ascorbic acid content following irradiation. In general, there were no changes in soluble solids, reducing sugars, sucrose, and total sugars. Irradiation increased pectinesterase activity and the water-soluble pectin fraction but decreased the versene-soluble and protopectin fractions.

Table 7 Anthocyanin Content of Loring Peaches

Stage and temperature	Storage, days	Anthocyanin content*			
		Unirradiated	108 krads	216 krads	432 krads
Mature green, 68°F	0	0.08	0.08	0.05	0.08
	6	0.09	0.11	0.14	0.43
	12	0.10	0.11	0.15	0.57
Firm ripe, 35°F	0	0.58	0.56	0.47	0.46
	7	0.37	0.49	0.39	0.40
	14	0.19	0.20	0.33	0.46

*A comparative value measured at A = 535 nm.

Respiration

Immediately after irradiation, evolution of CO_2 from fruit increased, but 18 hr later this activity decreased. The rate of CO_2 evolution then followed a similar pattern for both irradiated and nonirradiated fruit, with the respiratory rate for irradiated fruit greater.

MANGOES

Anthracnose infection of mangoes causes major losses of the fruit. Irradiation reduces the extent of the anthracnose infection and retards the rate of ripening.

Ripening

Mangoes were visually rated for ripening according to the area of yellow-colored skin. Table 8 shows the delay effect of irradiation on the rate of ripening of Zill mangoes. There were no differences in the rates of ripening if they were or were not hot-water dipped (7 min at 120°F) before irradiation.

Table 8 Effect of Irradiation on Ripening of Zill Mangoes

Storage (68°F), days	Dose, krads	Ripening rating*	
		No dip	Dip (7 min, 120°F)
5	0	8.3	7.3
	105	14.0	14.0
	157	15.3	14.7
	210	16.7	15.0
10	0	5.0	5.0
	105	7.7	9.0
	157	10.3	9.7
	210	10.0	10.7
17	0	5.0	5.0
	105	5.0	5.0
	157	5.3	5.7
	210	8.0	7.3

*Fully green = 25, fully ripe = 5.

Decay Control

Table 9 shows the significant reduction in anthracnose infection that was obtained by both irradiation and the dipping of fruit in 120°F water for 7 min before irradiation. When the storage period was 17 days, the reduction in infection was found only for those mangoes which received the higher irradiation doses—157 and 210 krads.

TOMATOES

Tomatoes are susceptible to Alternaria infections, and the infection is not controlled by irradiation doses that do not damage the fruit. Irradiation retards the rate of color development in tomatoes, and the treated fruit is not as firm. No significant differences occur in organoleptic preference ratings between irradiated and nonirradiated tomatoes.

Table 9 Effect of Irradiation on Anthracnose Infection in Zill Mangoes

Storage (68°F), days	Dose, krads	Infection rating*	
		No dip	Dip (7 min, 120°F)
5	0	14.3	9.7
	105	9.0	7.3
	157	8.3	6.3
	210	7.3	6.7
10	0	20.3	19.3
	105	12.3	7.3
	157	11.0	8.0
	210	10.7	9.3
17	0	25.0	25.0
	105	25.0	25.0
	157	18.7	13.0
	210	17.7	9.7

*Severe infection = 25, no infection = 5.

Decay Control

Tomatoes irradiated after the appearance of color on the blossom end developed more decay during ripening at 68°F than the nonirradiated fruit.

Ascorbic Acid

Tomatoes exposed to 300 krads contained less reduced ascorbic acid than nonirradiated fruits or those exposed to 100 krads (Table 10). However, the decrease in ascorbic acid at the higher dose was only about 10% of the acid present in the untreated fruit.

Table 10 Ascorbic Acid Content of Floradel Tomatoes Irradiated 3 Days After Color Break and Stored at 68°F

Storage, days	Ascorbic acid, mg/100 g			
	Unirradiated	100 krads	200 krads	300 krads
0	21.5	18.6	18.5	19.6
2	20.7	23.1	19.3	19.1
4	19.6	20.3	21.1	18.5
7	21.0	21.7	20.0	17.9

Respiration

The effect of irradiation on respiratory rate was influenced by the fruit maturity at the time of treatment. Irradiated mature green fruit showed an immediate increase in respiratory activity followed by a slight decrease in CO_2 evolution for about 18 hr. The maximum respiratory activity occurred 30 hr after irradiation. After 4 days, respiratory activity was similar to that of control fruit.

Fruit irradiated 1 day after color break showed an immediate increase in CO_2 evolution, but after 24 hr the respiratory rate was slightly below that of the control fruit.

Color

Fruit irradiated to 150 krads at color break showed a 2-day delay in reaching the orange-pink color as compared to nonirradiated fruit. No differences in rate of color development were observed between control fruit and fruit irradiated 2, 4, or 6 days from color break.

MICROBIOLOGICAL STUDIES

Stem-End Decay of Citrus Fruits

Phomopsis citri, stem-end decay, causes extensive losses of citrus fruits. Navel oranges were inoculated with spores and 16 hr later irradiated to 100, 200, or 300 krads. After 14 days at 68°F, the unirradiated fruit was starting to spoil but the irradiated fruit was sound. After 42 days, 88% of the unirradiated fruit was spoiled, but the irradiated fruit showed only initial evidence of spoilage.

Bearss lemons were inoculated at the stem scar with *Phomopsis citri* spores and 15 hr later irradiated to 50, 100, or 200 krads. The 200-krad treatment reduced the amount of spoilage markedly.

Brown Rot on Peaches

Monolinia fructicola causes most of the spoilage losses of peaches. Loring peaches at two stages of ripeness were inoculated with spores and after 14 hr irradiated to 108 and 216 krads. After 6 days, the amount of spoilage was much less with the green fruit but there was little difference after 9 days of storage. The 108-krad dose reduced the extent of spoilage considerably below that of the untreated fruit. The 216-krad dose gave much better control than the 108-krad dose.

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FRUIT AND VEGETABLE BIBLIOGRAPHY REVISED

The Isotopes Information Center has announced the publication and availability of a revised bibliography (over 1250 references) on radiation preservation of fruits and vegetables. The earlier version of this bibliography consisted of an initial report and two supplements. However, searching through three volumes has been inconvenient and, since research in this field has increased greatly since publication of the second supplement, the three earlier bibliographies have now been combined in this single, revised publication with material added from the holdings of the Isotopes Information Center and from a compilation of foreign references provided by R. S. Kahan and B. Gilbert of the Soreq Nuclear Research Centre, Yavne, Israel. There are, as before, an author index and a subject index. For the benefit of the many users of the three previous bibliographies, references that appeared earlier are identified not only with their sequential number in this bibliography but also with the number assigned to them in the earlier publication(s) plus a letter designating in which of the three volumes the reference occurred. Requesters should ask for: Radiation Preservation of Fruits and Vegetables: A Bibliography, F. E. McKinney (Ed.), USAEC Report ORNL-IIC-11 (Rev.), 1971. Copies are available free from the Isotopes Information Center, Oak Ridge National Laboratory, Oak Ridge, Tenn. 37830, for as long as the supply lasts, and copies are also available from the National Technical Information Service, Springfield, Va. 22151.

(FEM)

MARINE PRODUCTS DEVELOPMENT IRRADIATOR PROGRAM: IRRADIATED FRESH SHRIMP SHIPPING STUDIES

By J. D. Kaylor, E. J. Murphy, J. H. Green, and W. S. Hill, USAEC Report TID-25543, National Marine Fisheries Service, October 1970.

Supplementary Keywords: *food preservation; industry, food; seafood; food evaluation, subjective; source, gamma;* ^{60}Co.

The use of radiation to prolong the shelf life of fresh shrimp while retaining an acceptable product has been demonstrated in sensory evaluations conducted by Gloucester Fishery Products Technology Laboratory. A dose of 100 to 200 krads will extend shelf life by a week or longer.

This report presents results of shipping studies conducted between northeast Florida and the laboratory in Gloucester, Mass. Three shipping studies were conducted with white shrimp and three with brown shrimp. The shrimp were flown to Gloucester, usually within 1 or 2 days of being caught, irradiated, and returned to Florida for shipment back to Gloucester by commercial truck. Temperature patterns during the truck trip were recorded. In two tests, shrimp were sent to Louisiana State University (LSU) for duplicate sensory evaluation.

Doses tested during these trials ranged from 25 to 300 krads; both LSU and Gloucester evaluators agreed that 100- to 200-krad doses gave the best results. Shrimp that made the round trip of 3000 miles were not more adversely affected than the duplicate samples held at Gloucester. Conditions of commercial shipment apparently do not provide stresses great enough to be revealed by sensory evaluation. Total bacteria in irradiated shrimp seldom exceeded total bacteria in control samples, which is the opposite of what normally holds true for fish fillets. All tests but one clearly showed that radiation gave a commercially desirable shelf-life extension of at least 1 week.

The present system of cooling, usually storage in ice, used in interstate shipments by truck gave internal shrimp temperatures of less than 40°F. Modern shipping practices, therefore, are not expected to be a limiting factor in the distribution of irradiated shrimp. (FEM)

Section
IV
Isotopes and Radiation Technology

Process Radiation Development

New Aspects in the Field of Gamma-Irradiation Plants

By H. Christen*

Supplementary Keywords: *facility description; plant scale; source, gamma;* ^{60}Co.

Three Sulzer irradiation plants—a laboratory-scale research unit and two industrial plants with different capacities—are described here. Each of these ^{60}Co plants has certain interesting features that distinguish it from other similar installations.

"DOUBLE-CELL" LABORATORY IRRADIATION PLANT

A so-called "double-cell" irradiation plant is available for use in laboratory-scale research. This is a self-contained irradiation unit—i.e., it needs no additional radiation shielding (Fig. 1). The plant consists mainly of a lead container, in whose center the ^{60}Co radiation source is located. The container is penetrated by a rotating ring in which are installed two irradiation chambers, with a volume of about 3 liters each. One chamber is normally in the irradiation position, with the radiation source on both sides of the chamber. A timer is provided for each chamber to regulate the irradiation time.

The plant can be loaded with up to 10 thousand Ci of ^{60}Co to give a dose rate in the center of the irradiation chamber of 1.1×10^6 R/hr. Because the source is arranged in two vertical racks, source rods with higher activities can be inserted in the upper and lower positions of the racks. Thus the isodose lines run nearly parallel and vertical in the vertical plane, and the dose is homogeneous in this plane.

Two tube coils may be fitted at the outer sides of the source racks for irradiation of liquid or gaseous materials.

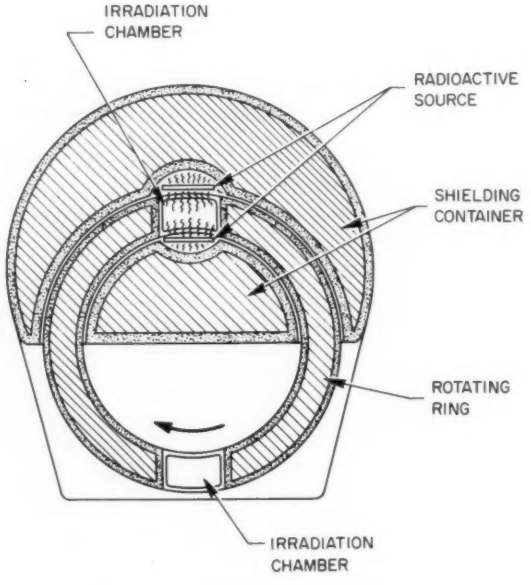

Fig. 1 Horizontal cross section of the Double-Cell Laboratory irradiation plant.

*Gamma-Irradiation Plant Group of Sulzer Brothers Limited, Winterthur, Switzerland. This article is a summary of one in *Sulzer Technical Review*, **52**(1): 59-63 (1970).

"UNICELL" INDUSTRIAL PLANTS

Two industrial plants designed particularly for irradiation of packaged goods are available. The material flow of the two plants—one for medium and one for large throughputs—is fundamentally the same (Fig. 2). After being completely packed in the production section, the items to be irradiated are stored on the conveyor for unirradiated products until they are automatically transported for sterilization. After irradiation, the items are deposited on the stacking conveyor for treated material, which is clearly separated from the one for unirradiated material. During normal working hours the items are forwarded to the dispatch department or to storage.

In selection of a Unicell model, the choice of size would be based on a number of factors, including throughput, product-density variation from one batch to another, and radiation utilization efficiency.

"Small Unicell" for Medium Throughputs

The Unicell for medium throughputs is a simple and economically favorable plant, which needs only a small capital investiment. In designing it, special attention was paid to a simple and uncomplicated mode of operation and to a plant arrangement that does not require much servicing.

The material to be irradiated is taken automatically from the first stacking conveyor and fed into the irradiation cell by a rotary cylinder. Thus the labyrinth passage is eliminated, resulting in considerable saving of space. The irradiation time is controlled by adjusting the conveyor speed. High operational safety is ensured by the absence, in the irradiation cell, of movable drive units or conveyor mechanisms that cannot be controlled from outside.

The maximum source strength is 200 thousand Ci of ^{60}Co. The source is lowered into its safe position in a lead container, which is also a transportation container. Thus a transfer from the container into the plant is avoided.

The concrete shielding consists of individual standardized shielding elements; this allows simpler and faster erection, as well as the possibility of dismantling the plant later for reerecting elsewhere if production increases to such an extent that a larger plant becomes advisable.

Because the material being irradiated makes only one pass on each side of the source, there is insignificant time loss when changing from one material to another that has a different density or requires a different absorbed dose and hence a readjustment of the irradiation time.

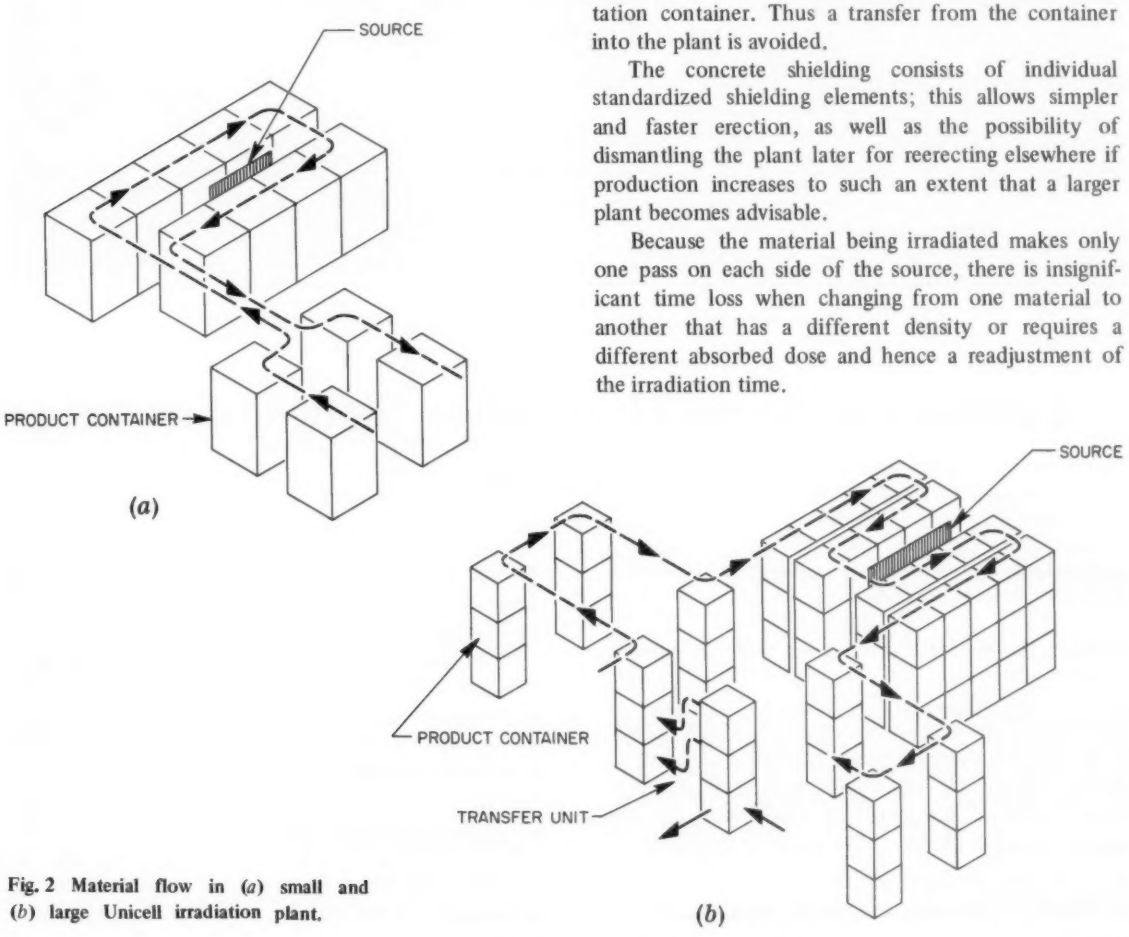

Fig. 2 Material flow in (*a*) small and (*b*) large Unicell irradiation plant.

"Large Unicell" for Higher Throughputs

The large Unicell was laid out for higher throughputs and, consequently, for source strength in the megacurie range. For higher utilization of the radiation, the plant was designed for several passes of the product around the source and at several levels in the irradiation cell. The number of passes depends on the density of the product.

This plant is provided with one or two labyrinth passages for supply and removal of the material, and the concrete for radiation shielding is cast as part of the building structure. The safe position of the source is generally in a pit below the floor, e.g., in a water pool, where the source can also be easily exchanged. Operation is fully automatic when the appropriate stacking conveyor system is provided. (MG)

RADIATION-INDUCED POLYMERIZATION. PART I, WOOD–PLASTIC COMPOSITES
AAEC-LIB-Bib-259, October 1970

RADIATION-INDUCED POLYMERIZATION. PART II, CONCRETE POLYMERS, AN IMPROVED TYPE OF CONCRETE OBTAINED BY USING RADIATION TO COMBINE CONVENTIONAL CONCRETE AND PLASTICS
AAEC-LIB-Bib-260, November 1970

RADIATION-INDUCED POLYMERIZATION. PART III, FORMATION OF POLYMER COATINGS ON SUBSTRATES
AAEC-LIB-Bib-261, November 1970

Three Australian reports compiled by E. A. Newland.

The three parts of the bibliography have approximately 150, 30, and 120 references, respectively. Sources of literature for the bibliographies were *Applied Science and Technology Index, Chemical Abstracts, Engineering Index, Nuclear Science Abstracts, Atomindex (IAEA),* and *Scientific and Technical Aerospace Reports.* Many of the references include either the *Chemical Abstracts* or the *Nuclear Science Abstracts* volume and abstract number. (REG)

Commercial Gamma-Irradiation Plant in Sweden

By C. G. Crawford*

Supplementary Keywords: *source, gamma; facility description; sterilization; industry, pharmaceutical.*

Abstract: *The Radona 215-kCi gamma-irradiation plant in Sweden is described.*

A commercial gamma-irradiation plant for sterilization of medical products is being operated in Sweden by Radona Irradiations AB at Skärhamn on the island of Tjörn, about 70 km north of Gothenburg. It was loaded with 215 kCi of ^{60}Co, supplied by the U. K. Atomic Energy Authority, in 1968. The plant was designed and built by H. S. Marsh Nuclear Energy Ltd. of Reading, England.

The Radona company manufactures, packages, sterilizes, and distributes medical products and sterilizes materials for customers under contract. This mixed load provides some special problems that do not occur with a plant handling only an in-house load.

PLANT DESCRIPTION

The plant (Fig. 1) is located at one end of a building in a 110- by 40-m area. The floor space not

*Radona Irradiations AB, 440 60 Skärhamn, Sweden.

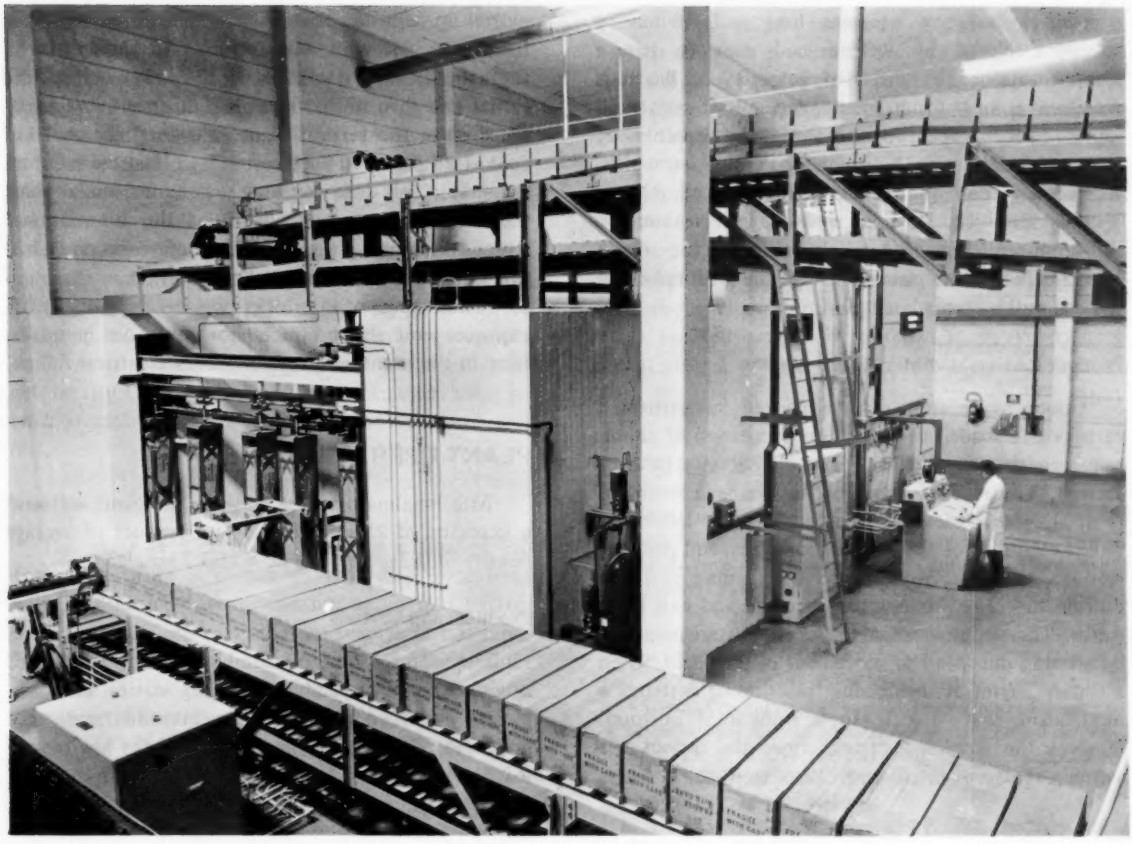

Fig. 1 Radona gamma sterilization plant.

occupied by the irradiation cell and its ancillary equipment is devoted to storage and quarantine areas for nonsterile and sterile goods. Input and output conveyors, each 80 m long, run down the center of the building on each side of a wall, which divides the "sterile" from the "nonsterile" area. Goods are delivered and dispatched through two large doors at the opposite end of the building from the cell, so that the flow pattern is down the nonsterile side, through the cell, and then out through the sterile area.

Cell

The cell, internally 5 by 5 by 3 m, is cast in steel-reinforced compacted concrete with a density of about 2.35 g/cm^3. The roof and walls are 1.65 m thick.

The entrance and exit labyrinths for goods are right-angled and of such a width and height that only the loaded vertical carriers can travel along them, leaving insufficient space for a person to enter. A pressure-sensitive footplate in the floor at the entrance, when stepped on, causes the plant to immediately revert to a safe condition, with the source shielded. The personnel door—a stepped steel box filled with concrete—runs on rails into an opening in the side of the cell and is fully interlocked with the source-raising mechanism.

Source

Shielding and cooling arrangements were designed for an ultimate source activity of 1 MCi. Above the source plaque is a stepped shielding plug. When not in use, the source is stored in a dry pit centrally placed in the floor of the cell.

The source frame is a vertical grid of parallel horizontal tubes. The source elements, with an initial activity of 12 to 22 kCi, are assemblies of 24 rods in

stainless-steel tubes, 660 mm long and 38 mm in diameter, closed by welded end caps to give a leakproof unit. The different activities of the rods facilitate source management and allow sequential replacement of decayed elements. The elements are delivered to the plant in a shielded container, which, when locked onto the loading face of the cell, releases a safety interlock. The source plaque is then raised to the appropriate position for insertion of the source elements, which are passed through the loading hole in the cell wall by articulated push rods.

Product Carriers and Monorail System

Goods to be sterilized are packed in corrugated cartons and loaded in an endless procession of aluminum carriers, 2 m high and 330 by 280 mm in cross section. The carriers travel on an overhead monorail (Fig. 2), which goes through the entrance labyrinth, passes each side of the source four times, and goes out through the exit labyrinth to the loading position, outside the cell, between the entrance and exit labyrinths. The radiation dose to the products is adjusted by altering the speed of movement of the chain. When a carrier arrives at the loading position, it actuates a mechanism that tilts it to a horizontal position, removes the sterilized cartons from the top of the carrier, replaces them with "half-sterilized" goods moved up from the lower half of the carrier, and loads the lower half with new cartons. All cartons travel through the cell twice, first in the lower half of the carrier and then in the upper half to ensure leveling of the dose in the vertical plane. The total accumulated dose at all points in each carton is at least the required minimum. This dose-averaging system in a similar plant was described in more detail at the International Atomic Energy Agency (IAEA) conference on Industrial Uses of Large Radiation Sources.[1]

A full range of interlocks protects personnel from exposure and also products from overdose or underdose in the event of a mechanical or electrical failure.

PLANT OPERATION

With maximum source activity, the plant will have a capacity of 25 tons per day of product of average density of 0.2 g/cm^3 at a dose of 3.2 Mrads.

Products and Cartons

The plant is used mainly for sterilization of single-use medical products, ranging in size and complexity from simple blood lancets and needles to peritoneal dialysis sets and tube sets for heart–lung machines.

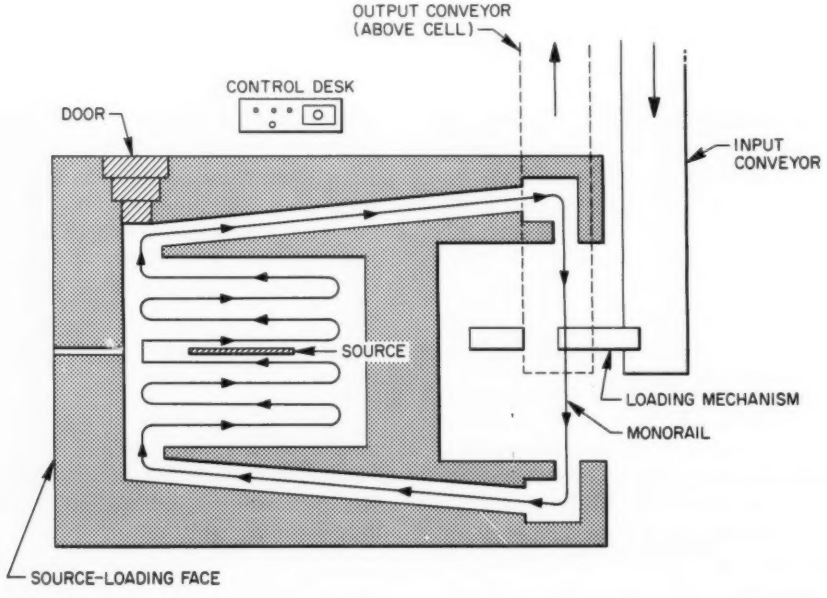

Fig. 2 Radona cell.

All products must be packed in cartons of size suitable for loading on the carriers and strong enough to withstand the thrust of the hydraulic ram that transfers them from the input conveyor to the carrier and from the carrier to the output conveyor. It is here that the special problems involved in running a service plant are encountered. Even though carton-strength and -dimension specifications are sent to all service customers, cartons are sometimes below the required standard, and, if filled with soft products, they will collapse. Often empty cartons are supplied to the customer to avoid trouble. This experience suggests that, when a plant is designed for sterilization of cartons from many customers, the tolerances must be much greater than when it is to be used for an in-house load.

Dose

The plant is normally run to give a dose of 3.2 Mrads. This dose is chosen so that after processing there is a probability of one nonsterile unit in 10^6 units processed, assuming that an average of not more than 50 organisms per unit are present on the product and that all these organisms are equal in radiation resistance to *Streptococcus faecium* A_2 1. This "safety margin" of 10^6 is based on the IAEA *Code of Practice for Radiosterilization of Medical Products,*[2] and 3.2 Mrads is regarded in the Scandinavian countries[3] as the minimum acceptable dose, although in other parts of the world 2.5 Mrads is accepted. The background to this method of expressing sterility requirements has been described by Christensen, Holm, and Juul[4] and by Kallings.[5]

The process is controlled by a presterilization count, daily dosimetry tests, and a continuous check with recording instruments to indicate normal operation of the plant and constancy of carrier speed. When a batch of goods is delivered for sterilizing, the average bacterial count per unit is determined. The batch is then passed through the normal 3.2-Mrad process and is held in quarantine until the bacterial count is completed (normally 7 days). If the average count is >50, the goods are returned to the cell for an additional dose, but this happens only infrequently. When it does, customers are urged to improve their manufacturing hygiene. Product sterility is checked after the irradiation only at the customer's request. A small random sample is unlikely to show anything useful after a process with a probability of failure of 10^{-6} (MG)

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3. E. A. Christensen, L. O. Kallings, and D. Fystro, Microbiological Control of Sterilization Procedures and Standards for the Sterilization of Medical Equipment, *Läkartidningen,* 66: 5117 ff (1969) (in Swedish); *Ugeskr. Laeger,* 131: 2123 ff (1969) (in Danish); *Tidsskr. Norske Laegforen.,* 89: 1806 ff (1969) (in Norwegian).
4. E. A. Christensen, N. W. Holm, and F. Juul, The Basis of the Danish Choice of Dose for Radiation Sterilization of Disposable Equipment, Danish Report Risö-140, September 1966.
5. L. O. Kallings, Review of the Code of Practice for the Radiosterilization of Medical Products, in *Radiosterilization of Medical Products and Recommended Code of Practice,* Symposium Proceedings, Budapest, 1967, pp. 433-444, International Atomic Energy Agency, Vienna, 1967 (STI/PUB/157).

TABULATED DOSE DISTRIBUTION DATA FOR GAMMA IRRADIATOR DESIGN

BNL-50147 (T-418), Revised, December 1970

AND

TABULATED DOSE UNIFORMITY RATIO AND MINIMUM DOSE DATA FOR GAMMA IRRADIATOR DESIGN

BNL-50145 (T-516), Revised, December 1970

By F. X. Rizzo, L. Galanter, and K. Krishnamurthy, Brookhaven National Laboratory.

These reports supersede BNL-50147 and BNL-50145, respectively, in which errors due to computer-program errors were discovered after the reports were issued. The errors, although minor for ^{60}Co sources, were significant for ^{137}Cs sources. The revisions contain values recalculated after correction of the code (GOLF P2). (MG)

Ezeiza Semi-Industrial Irradiation Plant*

Supplementary Keywords: *facility description; source, gamma; ^{60}Co.*

A semi-industrial irradiation plant was installed at Argentina's Ezeiza Atomic Center's High-Intensity-Radiation Source Division to carry out irradiation services and to provide technological and economic data for use in constructing special irradiators. Because the plant was designed to be flexible enough for use in various types of applications, its operating conditions are not optimum for any one process. The plant space (Fig. 1) is divided into an irradiation chamber; a room for the machinery that moves the source into and out of the storage pool and the product carrier around the source; a control-panel room; a storage depot; a preparation room; and office and record space.

The 6.40- by 12.00- by 4.00-m irradiation chamber has 1.80-m-thick walls and roof of ordinary concrete. Labyrinth openings in various parts of the walls allow entry of pipes and other services. The chamber, designed for use with 1 MCi of ^{60}Co, at present contains only 225 kCi. The ^{60}Co was obtained partly from Atomic Energy of Canada Limited (140 kCi) and partly from the Radiochemical Centre of England (85 kCi). Both the Canadian and the British products are doubly encapsulated in stainless steel, 2260 and 2500 Ci per capsule, respectively. The capsules are distributed among 30 stainless-steel cylinders, from which the source array is made up.

When an irradiation is to be made, the source is raised into the irradiation chamber from the bottom of the L-shaped pool. The material to be processed is loaded in the preparation room onto carriers on a 5-ton-capacity monorail. The carriers then go through a labyrinth, with a minimum cross section of 2 by 2 m, to the irradiation room, around the source at a rate regulated to give the desired dose, and back to the preparation room. (MG)

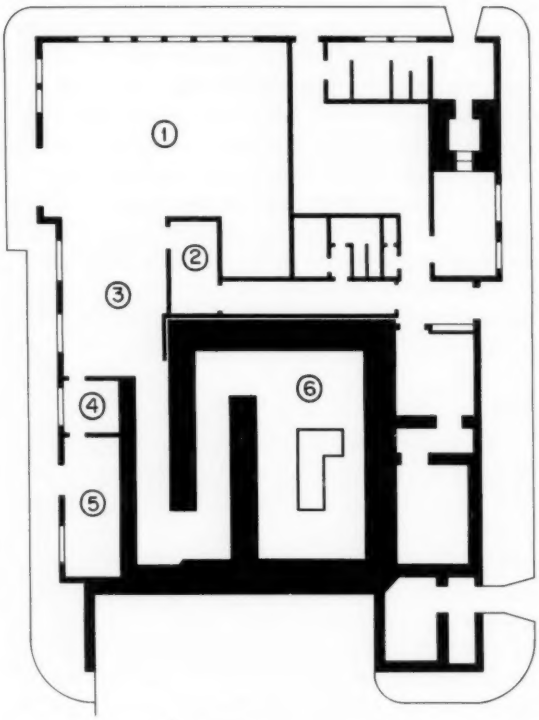

Fig. 1 Ezeiza semi-industrial gamma-irradiation plant: (1) depot; (2) office; (3) preparation; (4) control panel; (5) machinery; (6) irradiation chamber.

*Summary of report (in Spanish) by C. C. Papadopulos, National Atomic Energy Commission, Buenos Aires, Argentina, Report CNEA-272, 1970, 25 pp.

Section

V

Isotopes and Radiation Technology

Source Development

Recent Developments in Radioisotope Neutron Sources

By R. W. Tolmie*

Supplementary Keywords: *source, neutron; review, technical; comparison; conference;* ^{227}Ac, ^{210}Po, ^{228}Th, ^{242}Cm, ^{241}Am, ^{238}Pu, ^{88}Y, ^{124}Sb.

Abstract: *About 20 radioisotopes are used in making neutron sources. The (α,n) sources like ^{241}Am–Be and ^{238}Pu–Be are widely used in instruments and in other applications that require low-output, long-lived sources. Actinium-227, ^{210}Po, ^{228}Th, and ^{242}Cm with beryllium are available for higher outputs. Physically small, fast-neutron sources can be economically made from ^{252}Cf spontaneous fission sources, whereas the (γ,n) source ^{124}Sb–Be is more suitable for physically large very-high-output thermal-neutron sources. The (γ,n) sources and some special (α,n) source designs provide for switching the neutron output off and on.*

Until the last few years, by far the most familiar radioisotopic neutron sources were those which used the (α,n) reaction, utilizing ^{226}Ra, irradiated radium, ^{210}Po, ^{227}Ac, and other alpha-ray emitters with a beryllium target. These alpha-ray sources have now been largely supplanted by some of the transuranium isotopes, particularly ^{238}Pu and ^{241}Am, and (γ,n) sources like ^{124}Sb–Be are used when very high outputs are needed. Californium-252 is used when small point sources are needed. This article reviews the current status of these three types of sources.

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(α,n) SOURCES

Ansell and Hall[1,2] of the British Radiochemical Centre, in reviews of developments of (α,n) sources, point out that there are now about 20 nuclides to choose from, where once only the isotopes of radium and their daughters were available. Radium-226–Be was initially used in well logging but was superseded by ^{210}Po–Be, which was smaller, produced more neutrons, and presented less of a gamma-ray hazard. Subsequently, ^{239}Pu–Be and ^{241}Am–Be were used, followed by the more compact ^{238}Pu–Be sources, and now ^{252}Cf is coming into use for this application. However, the earlier sources are not completely obsolete. The gamma radiation from ^{226}Ra–Be is still used in some moisture meters for density determinations, and other meters similarly use ^{241}Am–Be sources, together with ^{137}Cs.

Usually (α,n) sources are made by preparing an intimate mixture of the source with beryllium metal to form a powder, pellet, or alloy. The principal developments in the manufacturing process have been improvements in encapsulation materials and sealing techniques, but there have been some requirements for sources of special shapes. One of these requirements is for an annular source designed to fit around a BF_3 counter in a soil-moisture meter. The source strength needed is 10^5 to 10^6 n/sec, obtained by adding 50 to 500 mCi of ^{241}Am to about 0.7 g of beryllium and pouring the powder mixture into a 0.25-in.-high annular ring, using an annular loading funnel. The capsule is sealed and then is again encapsulated in stainless steel. In a check for possible movement of the

powder, one such source was vibrated for 24 hr and also subjected to repeated dropping through a 1-in. distance for a similar period. There was no evidence of a change in or anisotropy of the output.

Somewhat larger sources, with outputs to 10^8 n/sec, have been used for neutron activation analysis, particularly for fluorine, utilizing (n,p), $(n,2n)$, and (n,α) reactions. There is possible interest in making such sources with central holes large enough to fit around a human hand or foot for the measurement of calcium in bone.

Another encapsulation problem arises from the need to produce line sources having uniform activity per unit length for industrial gages and interstitial and intercavity neutron therapy. The Radiochemical Centre has developed a rotary swaging technique for producing such sources. A mixture of $^{241}AmO_2$ and beryllium-metal powder is loaded into a ductile aluminum tube, which is sealed with end plugs and then extended into wire form in the swaging machine. The wire is cut to the desired length and sealed at the cold sheared ends. The enclosed mixture is very dense and adhesive after being swaged. The variations in neutron output along the length can be less than ±10% between 1-in. segments.

Some preliminary work has been done by Ansell and coworkers on the production of needle sources suitable for clinical therapy. Curium-242 was chosen because of the need for high specific activity. One curie, as 0.3 mg of the oxide, plus 10 times that mass of beryllium was loaded into a platinum inner capsule (0.85 mm in inside diameter, 1.25 mm in outside diameter, and 15 mm long), which was sealed by gold brazing. This was then inserted into a Pt–Ir sheath and sealed by resistance welding. The output was 3×10^6 n/sec, but it was estimated that this could be increased to 10^7 n/sec.

Work has also been carried out on neutron sources capable of withstanding high temperatures. Above 600°C, beryllium will attack stainless steel, and, if the capsule should be ruptured, the powder content might cause severe contamination. A development study using $^{241}AmO_2$ in a high-temperature sintered compact with BeO was undertaken. The BeO target requires that the activity be doubled, but the yield per unit volume remains about the same as with the beryllium metal because densities of around 2.6 g/cm^3 can be achieved, as compared to 1.3 g/cm^3 for the metal mixture. The sintered pellets did not powder in pressure tests but ultimately broke into discrete pieces. From pellets containing 10 mCi of ^{241}Am immersed for 8 hr in water at 50°C, the amount leached out was less than 0.05 μCi.

A switchable neutron source was developed to serve as a calibration standard in a neutron dosimeter. It consists of half shells of an alpha-emitting foil and of beryllium attached to a button magnet and free to rotate within a sealed stainless-steel capsule. An external magnet rotates the half shells to align them with a fixed pair of similar half shells, so that neutrons are produced when the alpha foils are opposite the beryllium targets. With 5 mCi of ^{241}Am, an output of 10^4 n/sec was obtained in the "on" position and 1.5×10^2 n/sec in the "off" position.

A somewhat similar source, described by Jordan, Carew, and Barkley,[3] is designed to produce a continuously variable neutron output by moving ^{210}Po-coated and beryllium arrays of plates in apposition to each other. The ^{210}Po, entrapped in ceramic microspheres, is coated on aluminum plates and held there by a thin epoxy film. Three millicuries was applied to a source area of 6.25 cm^2, using 1 Ci/g microsphere material having a bulk density of 2 g/cm^3. The energy of the alpha particles decreases as they pass through the ceramic and epoxy materials, especially at angles other than normal to the support, so that the alpha output is effectively cut 50%. The output from the test source was 111 n/sec per square centimeter of source area, and, with parameters adjusted to the optimum, was calculated to be 3×10^3 n/sec per square centimeter. This output is comparable to what might be obtained with a ^{210}Po metal–Be assembly in which the source is covered by a thin window to contain the polonium.

For applications requiring high neutron output per volume—e.g., for clinical treatment needles—it appears likely that ^{252}Cf will prove more suitable than (α,n) sources. Californium may also be more suitable for reactor startup because of its low mass and the fact that it does not require beryllium, which raises problems of helium production under irradiation. For applications in which these special features are not important, (α,n) sources will continue to be used, especially where long-lived sources are needed. Ansell and Hall[1,2] note that, of the three preferred (α,n) sources, ^{244}Cm–Be is not generally available, and, since ^{238}Pu is subject to many restrictions because it is a fissile material, ^{241}Am may continue to be the most important nuclide, at least for low-intensity sources.

In the United Kingdom, ^{210}Po, ^{228}Th, and ^{242}Cm are preferred for making sources with outputs in excess of 10^8 n/sec, the trend being to the latter two. These sources are produced by reactor irradiation of ^{226}Ra and ^{241}Am and are primarily used in

reactors and subcritical assemblies where a longer-lived source is needed. Large power reactors, where a short-half-life source is acceptable, usually rely on the cheaper ^{124}Sb–Be (γ,n) sources.

Ansell and Hall summarize the properties of 11 types of (α,n) neutron sources and briefly discuss the choice of targets, theoretical and practical yields, and properties of the alpha-emitting nuclides and compare some of the neutron spectrums. Curium-242 and ^{228}Th present special production problems because of their parents being alpha emitters, a situation that results in extra heat from the alpha decay and pressure buildup from the production of helium and fission-product gases in the reactor. A typical target for production of ^{242}Cm is 400 mg of ^{241}Am as oxide homogeneously dispersed in nickel powder and cold compacted into pellets. The pellets are sealed in an inner capsule with a helium atmosphere. Then a second encapsulation is used, which also encloses a ^{226}Ra source whose radon emission is used for leak testing. The neutron irradiation lasts 3 weeks at a flux of 10^{14} n/(cm^2)(sec), producing 150 Ci of ^{242}Cm. The capsule is opened, the contents are dissolved in nitric acid, and the americium and curium are extracted by a combination of classical chemical methods and ion exchange. The americium is removed from the curium by oxidation with persulfate.

The ^{228}Th is produced by irradiating 3 g of radium carbonate for 9 months at 10^{14} n/(cm^2)(sec) to produce 40 Ci. The ^{228}Th is extracted by ion exchange. When eluted, it is free from its radium daughter. Several weeks are required to reach maximum neutron emission, and thus the radiological dose problems are eased for the operators in making the ^{228}Th–Be source.

Considerations in designing the (α,n) source capsules are discussed in Ref. 2. The ^{228}Th content is measured by determining the gamma emission from the source once it has grown into equilibrium. The strength of both types of sources is also measured by determining their heat output with an adiabatic calorimeter. The containers required for such sources are very large, and shipping regulations often require that the source capsules meet the "special form" requirements of the International Atomic Energy Agency (IAEA).

De Troyer, Droissart, and Poskin[4] have revived the use of ^{227}Ac for (α,n) neutron sources. Such sources have a modest gamma-ray output, a long (22-year) half-life, and a high output per unit volume. They report that production of large quantities is under way. The ^{227}Ac is extracted from irradiated radium in nitrate form and is precipitated as $Ac(OH)_3$ in a water–alcohol mixture. This gives a mean particle size of 0.2 μ. Then 325-mesh beryllium powder is added in a ratio of 10 : 1, and the mixture is ground, pelletized, and loaded into a capsule having a void volume large enough to cope with the helium buildup. A small vent is left in the capsule, and the capsule is heated to 900°C for several hours in an inert atmosphere to degas it and to transform the actinium hydroxide to oxide. The capsule is then welded, leak tested, and put into an outer capsule, usually made of type 304L stainless steel. Tests on such capsules indicate that they pass the IAEA "special form" requirements. The neutron yield is 85% of the theoretical value of 2.62×10^7 n/sec per curie after the 3 months needed to reach equilibrium. The gamma and bremsstrahlung dose rate is 180 mrads/(hr)(Ci) at 1 m. The cost of these sources is estimated to be $40 per 10^6 n/sec for sources having an emission of 10^7 n/sec and higher. When such a source is used in a reactor, the neutron output is increased by conversion of some of the ^{227}Ac to 2-year ^{228}Th, which is also an alpha emitter.

Wahlgren and Stewart[6] discuss the possible uses of ^{244}Cm–Be neutron sources. Within a decade, ^{244}Cm will become readily available as a by-product of power-reactor fuel reprocessing. A major attraction is its 17.6-year half-life. A test source was made up having a volume sufficient to contain 1 g of ^{244}Cm in a mixture with a Be–Cm atom ratio of 10 : 1. Aluminum was chosen as the capsule material because of its low neutron-capture cross section and ease of manufacture, and the capsule was closed with a powdered aluminum plug, which allows helium to leak out. The beryllium powder was carried through the curium hydroxide precipitation step to avoid losses in a grinding and mixing step. Gamma-ray-spectrum measurements showed only lines of ^{239}Np, produced in the decay of ^{243}Cm (0.05% abundance).

For practical evaluation of the ^{244}Cm–Be source, it was mounted in a variable-configuration moderator and used for neutron activation analysis of a variety of materials. A ^{252}Cf source was also used for comparison, and the results were normalized to a source output of 10^8 n/sec. Sensitivities were greater with ^{244}Cm–Be, because of the higher thermal-neutron yield, than with ^{252}Cf when small solid samples and (n,γ) reactions were studied. The difference was less significant with solutions and slurries. A factor-of-10 improvement is achieved for (n,p) or (n,α) reactions because of the high energy content of the neutron spectrum as compared to ^{252}Cf. The cost of ^{244}Cm–Be sources, when they become available, is estimated at about $1800 for a 2×10^8 n/sec source.

Turning to (γ,n) sources, it is important to realize that, for a reasonable neutron yield, such sources must use thick beryllium targets, which may be 20 cm or more in diameter. Tables of comparative source performance are frequently based on the use of rather thin beryllium sheaths, so that the indicated yields may be an order of magnitude less than what may be readily achieved in practice. Physically large sources are plainly of limited use in some cases, but they are ideal for activation analysis because they provide large volumes of uniform fluxes.

O'Brien and Schillaci[7] have described the potential for production of a variety of neutron-source nuclides in the Los Alamos Meson Physics Facility (LAMPF), which is to start operation in January 1973. The most interesting of these nuclides is 107-day ^{88}Y. The LAMPF is an accelerator that will accelerate protons to an energy of 800 MeV, with a beam current of 1 mA. The residual beam reaching the main beam stop is estimated to be 0.5 mA of 700-MeV protons, and plans are to use this excess beam for the production of radionuclides. Six target stations will be provided, with target thicknesses up to 2.5 cm being acceptable. The principal mechanism for radionuclide production is spallation. With a zirconium target, the cross section for ^{88}Y reactions is 45 mb, and the ^{88}Y gamma-ray energy is 1.836 MeV (99.5%). The calculated yield for a 4.5-cm-thick target and a half-life irradiation is 210 Ci. Data are presented for nine other radionuclides, but these all have some disadvantage, e.g., an inconveniently short half-life or a low abundance of the high-energy gamma rays. Since these sources provide a variety of neutron energies, they should be useful for energy-dependent applications. One LAMPF target station, if used full time for the production of ^{88}Y, could produce about 240 Ci/month, which limits the potential to rather low-intensity (10^7 to 10^8 n/sec) sources.

This article and the one by Kull, Schillaci, and Beyster[8] compare the ^{88}Y–Be source to ^{124}Sb–Be sources. For the small- to intermediate-size sources considered, their estimate of the cost of ^{124}Sb is $30 per curie,* at which the ^{88}Y is estimated to be competitive if its longer half-life is taken into account. In a nuclear-materials assay system, some advantage derives from the 200-keV neutrons of the ^{88}Y–Be as compared to the 27-keV neutrons from ^{124}Sb–Be, resulting in a greater degree of neutron penetration into the sample. An interesting feature of most of the (γ,n) sources is that the most energetic neutrons produced have energies less than threshold values of fast-neutron reactions—(n,α), (n,p), or $(n,2n)$. With only (n,γ) reactions to consider, the untangling of spectrums in neutron activation analysis is considerably simplified. There are circumstances in which it is preferable to use fast-neutron rather than thermal-neutron reactions, and in such cases the sample can be surrounded by a thin foil of ^{235}U, which very effectively shields the sample from thermal neutrons so that a flux of fast neutrons is produced. The neutron spectrum is a fission spectrum similar to that obtained with ^{252}Cf.

One point not generally realized about (γ,n) sources is that for large sources the shields can be much smaller and lighter than those for (α,n) or ^{252}Cf sources. This is of course the opposite of the situation that obtains for small sources and is due to low-energy neutrons, particularly those from ^{124}Sb–Be, being readily thermalized and absorbed. Thus the shield is primarily composed of about 35 cm of lead for the gamma rays from the antimony and prompt gammas produced around the source. Assemblies have been made that will hold more than 10 thousand Ci. At a yield of 2×10^7 n/sec per curie, the output is 2×10^{11} n/sec, which is considerably greater than that of any other type of radionuclide neutron source yet made.

Currently, the greatest interest is centered on the radionuclide ^{252}Cf, from which high-output point sources can be made and which has a convenient (2.65-year) half-life. McDonell et al.[9] describe the preparation of such sources, about 50 of which have been made to date at the Savannah River Laboratory. They range in size from 5 μg to 5 mg or more and emit 2.34×10^9 n/sec per milligram due to spontaneous fission. The californium oxide sources are encapsulated in a 90% Pt–10% Rh alloy inner capsule, which is enclosed in a type 304L stainless steel or Zircaloy-2 outer capsule. The Zircaloy-2 minimizes interfering gamma emissions in prompt-gamma activation-analysis applications.

The sources are prepared from californium oxalate, precipitated from 0.1*M* HNO_3 solution by the addition of excess oxalic acid, onto a porous type 316 stainless-steel or platinum-alloy disk in the inner source capsule. About 500 μg of terbium, in nitrate form, is added as a carrier for sources weighing less than 500 μg. The precipitation is complete in 1.5 hr. Another porous metal disk is placed over the source, and the oxalate is dried for 1 hr at 200°C and calcined for 30 min at 700°C to convert it to oxide. The inner

*For large sources, the cost of ^{124}Sb is less than $1 per curie.

capsule is then sealed with tungsten–inert-gas-welded end plugs, heated 15 min at 1000°C, pressurized in helium at 300 psi for 60 min, and leak tested. The sealed capsule is decontaminated ultrasonically, with alternate washings in 1*M* HNO_3 and distilled water, until 1 ml contains less than 50 dis/min alpha and 200 counts/min beta–gamma contamination. The outer capsule is sealed with a welded end plug, helium leak tested, and ultrasonically decontaminated to a level of less than 9 dis/min alpha and 10 counts/min beta–gamma transferable activity as indicated by a wipe test. The output is measured with a fission counter.

In a typical series of five 100-μg source preparations, the yields ranged from 94.0 to 99.7% with a terbium carrier. For a 1-mg source prepared without a carrier, the yield is typically 92%. For the two 5-mg ^{252}Cf sources that have been prepared, the yield was greater than 98%. The ^{252}Cf lost to the filtrate and wash solutions was largely recovered.

The isotopic purity of the early sources has varied considerably—45% ^{252}Cf in one series and 86% in another. The ^{249}Cf content can reach around 24% and the ^{250}Cf, 25%. The ^{251}Cf varies from 3 to 6%. A fairly wide variety of chemical impurities typically range four to five times the ^{252}Cf content. Objectives set for these impurities are

Organic degradation products: essentially none.
Other total solids: maximum of four times the californium concentration.
Actinides–lanthanides other than californium: maximum equal to californium concentration.

The sources were handled in hot cells through the manufacturing steps. The 5-mg sources, after fabrication, were each suspended at the midpoint of a cord held at each end by an operator. The source was drawn up through an access port in the cell roof slab and deposited in a "Benelex" cask. The operators, 20 ft away from the source, were momentarily exposed to a dose rate of up to 1000 mrems/hr, principally of neutron radiation. At 5 in. from the cask surface, the dose rate was 100 to 250 mrems/hr.

McDonell et al.,[9] after examining the question of stability of the ^{252}Cf oxide, predict that there will be no deleterious incompatibility of the californium sesquioxide with the platinum-alloy capsule at temperatures up to 1000°C. The $^{252}Cf_2O_3$ could react with noble-metal oxides only until the limited amount of oxygen in the capsule is used up.

The heat produced from alpha decay and fission of ^{252}Cf is about 40 W/g. A capsule loaded with 10 mg of ^{252}Cf and losing heat by natural air convection would rise in temperature to 50°C above ambient in the inner capsule and to 34°C above ambient in the outer. Enclosed in a 1-ft thickness of a good insulator, the outer-capsule temperature rise was calculated to be 74°C.

Helium produced by alpha decay is the principal gas generated in the source, fission gases being less than 1% of the total. Complete alpha decay would produce about 88 cm^3 of helium (STP) per gram of ^{252}Cf. After 10 years' decay, the internal gas pressure in an inner capsule initially containing 10 mg of ^{252}Cf at 1 atm would be 89 psi at 0°C and 413 psi at 1000°C. If a leak released the gas to the outer container, the corresponding gas pressures would be 59 psi at 0°C and 275 at 1000°C. The calculated wall strengths indicate that the inner Pt–Rh capsule should withstand up to 6500 psi and the stainless-steel outer capsule 2400 psi for short times at 1000°C, with the Zircaloy-2 capsule being able to cope with more than double this value. Tests on prototypes showed that practical sources would realize these calculated figures.

Kok, Artigas, and Ray[10] further investigated the effects of source-encapsulation materials on the thermal-neutron flux from ^{252}Cf sources, using water as the moderator. Two sources were compared—one, used by the Battelle Memorial Institute, encapsulated in stainless steel and Pt–Rh alloy; the other, owned by the Oak Ridge National Laboratory, encapsulated in aluminum. Source and capsule dimensions were different for the two sources. The thermal fluxes at various distances from the source in water were calculated, using DOT–IIW code, which solves the two-dimensional Boltzmann transport equation with general anisotropic scattering by using a diamond difference-solution technique. The energy range was divided into 15 energy groups, of which the fifteenth corresponded to the thermal-energy range. All runs were normalized to a total source strength of 1 n/sec.

The fluxes from these sources were measured experimentally, using dysprosium foils, 0.25 in. in diameter and 0.001 in. thick, mounted on 0.05-in.-long rods of polystyrene, which does not become radioactive and has cross sections closely approximating those of water. The rods were stacked in a polystyrene tube placed against the source. After activation, a 60-min cooling time allowed the ^{165m}Dy to die away before the 0.0948-MeV decay gamma ray from ^{165}Dy was counted. The absolute flux was determined by activating gold foil at a distance of 0.563 in. from the surface of the source. The counting system was calibrated against gold foils activated at the National Bureau of Standards.

The magnitude and shape of the calculated thermal-flux distributions up to about 1.5 in. from the source were the same as for the experimental data. At greater distances, the calculated fluxes decrease more rapidly than the measured ones, apparently because of limitations in the computer code. The source in the aluminum capsule gave a 25% higher value at the peak normalized flux. Consequently it was recommended that a high-strength low-cross-section material, such as zirconium, be used for encapsulation to achieve optimum peak thermal flux.

Currie, McCrosson, and Parks[11] have suggested that the simple moderator might be replaced with a subcritical multiplying system. They used discrete ordinate codes to calculate neutron fluxes in $^{233}U-H_2O$ and $^{235}U-H_2O$ solutions containing a central ^{252}Cf source. Spherical solutions with $K_{eff} = 0.98$ could enhance the flux by factors of 6.4 and 4.8, respectively. The calculations were extended to cylindrical multiplying systems more directly like practical systems, but, for useful flux-enhancement factors, the assembly must be so close to criticality that it might be necessary to carry out safety procedures and meet regulations little different from those for a full-fledged nuclear reactor. At least one type of nuclear reactor now in operation, the Slowpoke, was designed to have an inherent negative temperature coefficient without the use of electromechanical devices so that it might be operated without continuous monitoring, etc.

For a cost comparison of all these different types of neutron sources, many factors must be considered, including half-life, encapsulation and shipping costs, decay during shipment, and interest on investment in very-long-lived sources. Heinrich[5] has developed expressions that take these factors into account so that, for a given set of conditions, a systematic cost comparison can be made of the average annual cost over a 10-year period. It is assumed that, after 10 years, either deterioration or obsolescence will be a limiting factor. However, a considerable degree of caution must be exercised in drawing conclusions from such calculations. For example, Heinrich's conclusion that ^{252}Cf sources are the most economical for outputs greater than 7×10^8 n/sec is true only for point sources. If physically large sources are acceptable (and they are, in fact, usually preferable for neutron activation analysis), $^{124}Sb-Be$ sources are less expensive on an annual cost basis for high-output systems. The cost per curie drops to less than $1 for large ^{124}Sb sources, which are very easy to produce, and thick beryllium targets can yield up to 2×10^7 n/sec per curie, which is much higher than the thin-target value chosen for the calculations. (MG)

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Section
VI
Isotopes and Radiation Technology

Isotopic Heat and Power Development

Comparison of Radioisotope-Fueled Energy Sources for a Prosthetic Device

By Joel D. Hixson*

Supplementary Keywords: *energy source; thermoelectric system; thermionic system; nuclear battery; worldwide; heart;* ^{238}Pu; ^{147}Pm.

Abstract: *Cardiac pacemakers fueled with* ^{147}Pm *and* ^{238}Pu *and using thermoelectric, thermionic, and betavoltaic energy conversion are compared.*

Radioactive decay has long been considered a potential source of energy for remote-system power requirements. Today the most challenging remote system being considered is the interior of the human body, and the power requirement is for an electronic prosthetic device. The cardiac pacemaker—the most common of these prosthetic devices—is the first such device to use a radioisotope for the power supply.

Approximately 50 thousand pacemakers were sold throughout the world in 1970, and their use increases each year. Currently used devices are powered by chemical batteries, which deliver an electrical pulse to the heart when the heart's normal conduction system is blocked. From a subcutaneously implanted pulse generator, containing the electronics and power supply, leads are connected to the heart either through a vein or the chest wall. There are two basic types of cardiac pacemakers—asynchronous devices, which deliver electrical pulses to the heart at a fixed preset rate, and synchronous or demand units (Fig. 1), which sense cardiac activity and pace the heart only when needed. Ten years of experience with the demand units has shown that the chemical battery is the limiting factor on a unit's life and reliability.[1] For this reason radioisotope-fueled electrical generators are being considered for use in implantable pacemakers.

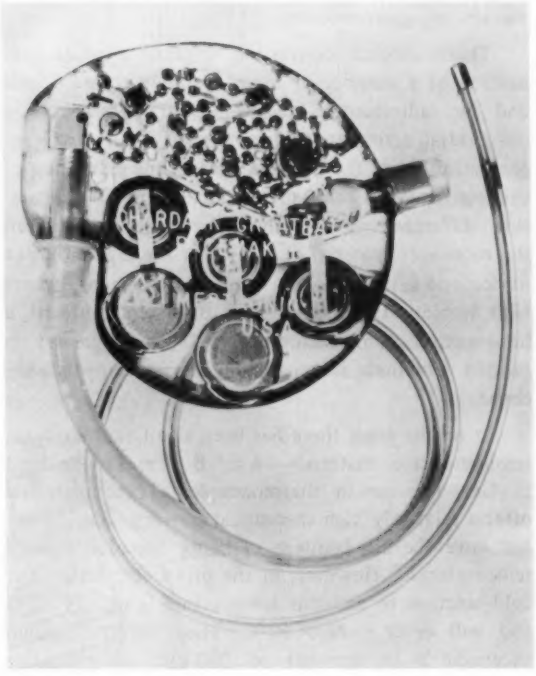

Fig. 1 Medtronic's model 5842 pacemaker with transvenous endocardial leads.

The power requirement for today's pacemakers is approximately 200 μW at 5 V. Radioisotopic genera-

*Medtronic, Inc., 3055 Old Highway Eight, Minneapolis, Minn. 55418.

tors of the microwatt size are being developed by many groups throughout the world, some of which were discussed in *Isotopes and Radiation Technology* 7(2) and updated in 8: 360-362. These generators convert the radioisotopic decay heat to electricity by one of three techniques: (1) thermoelectric, (2) thermionic, or (3) betavoltaic. This article compares systems using the three types of converters and different isotopic fuels.

THERMOELECTRIC CONVERSION

Larger electrical generators incorporating thermoelectric energy conversion were used in space and marine applications as early as 1960. Because of this history, most of the microwatt-sized generators today use thermoelectric energy conversion. The governments of the United States, United Kingdom, and France have sponsored programs to develop isotopic power suppliers for pacemakers, all of which utilize thermoelectric energy conversion.

Thermoelectric conversion requires a series connection of a number of thermocouples heated at one end by radioisotopic decay heat. The temperature differential across the thermopile results in a voltage generation from the Seebeck effect; the efficiency of this conversion is primarily a function of the temperature differential across the thermopile for a given thermocouple material (Fig. 2). For the higher-output devices used in space or marine applications, where high ambient temperatures are often encountered, a hot-junction temperature of 500°F or greater is needed. For these reasons metal thermoelements were chosen.

In recent years there has been significant study of semiconductor materials—e.g., Bi–Te, Pb–Te, and Si–Ge—for use in thermocouples. These materials offer a relatively high theoretical conversion efficiency but have the disadvantage of being degraded at high temperatures.[2] However, in the prosthetic device, the cold-junction or ambient temperature is usually 37°C and will never exceed 50°C. Thus, for the cardiac pacemaker's requirement of 200 μW, the optimum temperature differential for the semiconductor materials is only about 50°C, and Bi–Te (Fig. 2) appears to be the best thermocouple material in this application.

Bismuth telluride thermoelectric devices are being used in both the French and British programs to develop an isotopic power supply for cardiac pacemakers (see p. 222 of this issue). The French and British devices are very similar, and both use ^{238}Pu as fuel. The British device, designed by the Atomic

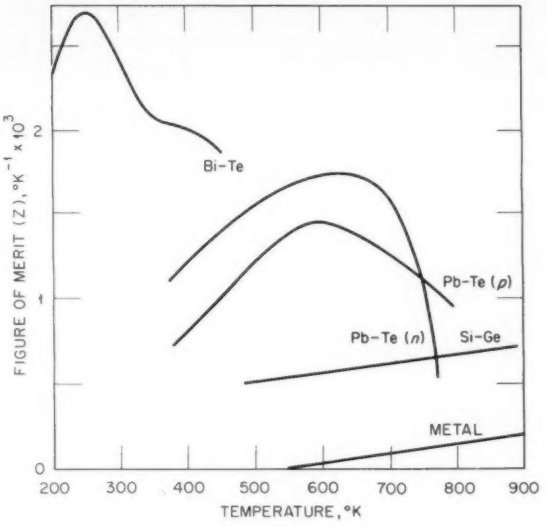

Fig. 2 Theoretical conversion efficiency of various thermocouples as a function of hot-junction temperature.

Energy Research Establishment (AERE) at Harwell, has a single container encapsulating 180 mg of plutonium nitride purchased from the USAEC. The thermoelectric unit, 17 mm in diameter and 49 mm long, delivers 400 μW of electric power at 0.4 V (Fig. 3). The thermal insulation includes a vacuum. Such generators, incorporated into asynchronous pacemakers manufactured by Devices Implants Ltd., were implanted in two patients in the summer of 1970. One device was subsequently removed after a period of about 3 months with power-supply malfunction.[3] It has been said—but not reported in the literature—that the second device has also been removed.

The French unit (Fig. 4), manufactured by Société Alcatel in Paris, is fueled by 142 mg of a Pu–Sc metal alloy.[4] The fuel is doubly encased—in both a tantalum and a platinum capsule. The device, which is 23 mm in diameter and 40 mm long, delivers 250 μW at ~0.5 V and uses a microporous thermal insulation at atmospheric pressure.[5] This power supply has been incorporated into a ventricular-programmed-demand pulse generator (Fig. 5) manufactured by Medtronic. Thus far, four of these pacemakers have been implanted in patients in France, the first[6] on Apr. 27, 1970. All pacemakers continue to function normally.

Other Bi–Te thermoelectric power supplies are in the developmental stage at Siemens Company in Germany and at Gulf Radiation Technology, a division of Gulf Energy & Environmental Systems Inc.; 3M

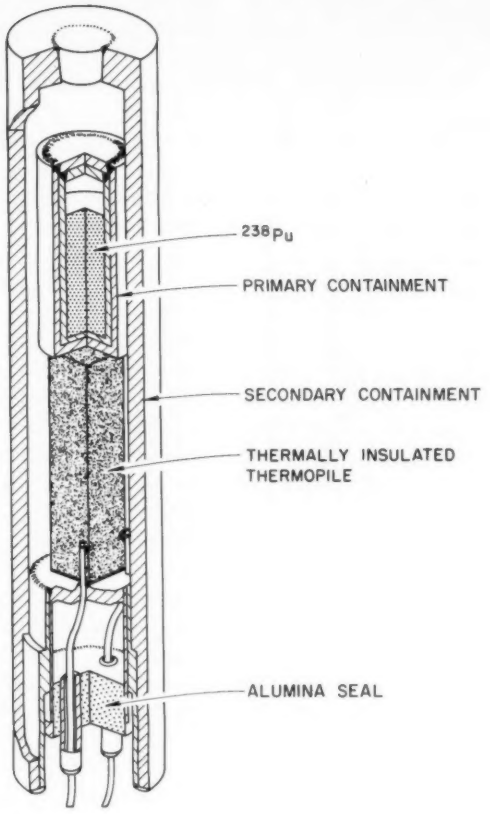

Fig. 3 The United Kingdom Atomic Energy Authority's ^{238}Pu energy source, built by the Atomic Energy Research Establishment at Harwell, England.

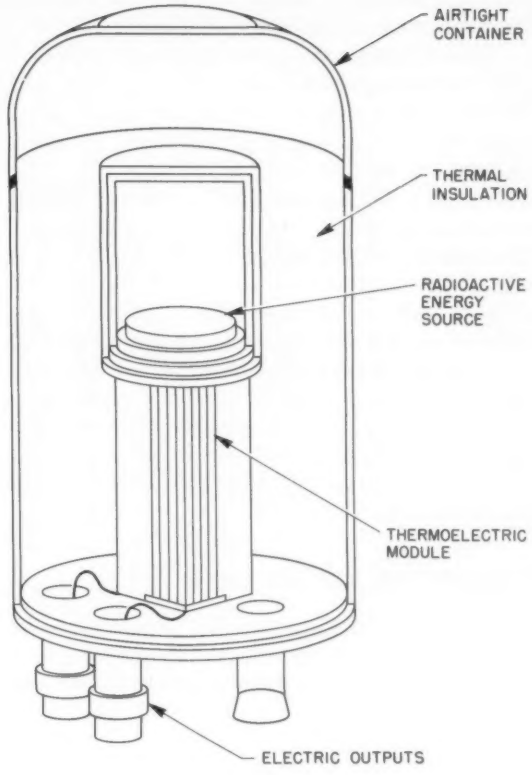

Fig. 4 Generator built by Société Alcatel of Paris.

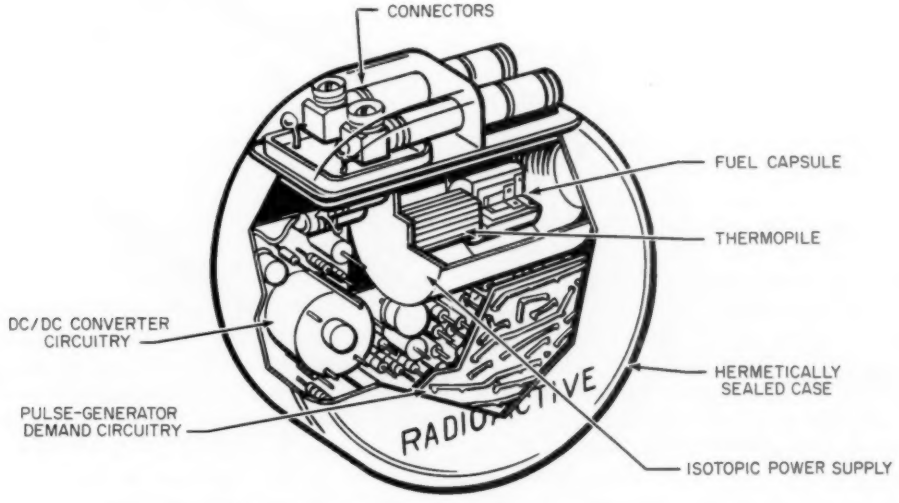

Fig. 5 Medtronic's Laurens–Alcatel model 9000. Powered by Alcatel's ^{238}Pu battery.

Company; Donald W. Douglas Laboratories; and Hittman Associates, Inc., in the United States.

Most of today's interest in developing microwatt-sized radioisotope-fueled generators is a result of a USAEC contract initiated in 1964 to develop a power supply for cardiac pacemakers, the prime contractor for which is NUMEC (Nuclear Materials and Equipment Corp.). The contract specified the use of metal thermocouples (Cupron Special and Tophel Special). The NUMEC design contains 500 mg of ^{238}Pu as an oxide triply encapsulated in Ta–10% W, Hastelloy alloy C, and titanium. Interwoven in a vacuum-insulated package are 528 thermocouples, each 0.6 by 5 by 2.8 mm (Fig. 6). This device delivers 240 μW at ~6 V and has been mated to asynchronous pacemaker circuitry supplied by Cordis Company of Miami, Fla. Eight of these units have been implanted in eight dogs[7] during the last 18 months, but six have been removed due to electronic malfunction.

THERMIONIC CONVERSION

The second energy-conversion technique suggested for microwatt power sources is thermionic. Figure 7 shows a ^{238}Pu-fueled thermionic device, the Isomite, built by Donald W. Douglas Laboratories of Richland, Wash.[8]

Thermionic devices have three main parts: (1) the emitter, (2) the working fluid, and (3) the collector. The emitter is the outer surface of the encapsulated radioisotopic heat source. The heat input imparts sufficient energy for electrons to overcome retarding forces at the surface of the metal and escape. The so-called "working fluid" in this converter is an electron gas or partially ionized plasma. The electrons from the emitter pass across the working fluid, overcome retarding forces, and migrate to the collector where part of their energy is lost in the form of heat and the remainder drives electrons through an external load back to the emitter.[9] As with thermoelectric conversion, almost all thermionic design in the past has been on larger power systems. As output power decreases in the thermionic devices, the optimum emitter temperature decreases, and, since the efficiency of this type of conversion is largely a function of the emitter temperature, thermionic devices in the low-microwatt power range may be impractical. However, because of this property, a given thermionic design has the capability of operating over a wide range of power requirements simply by increasing the fuel inventory

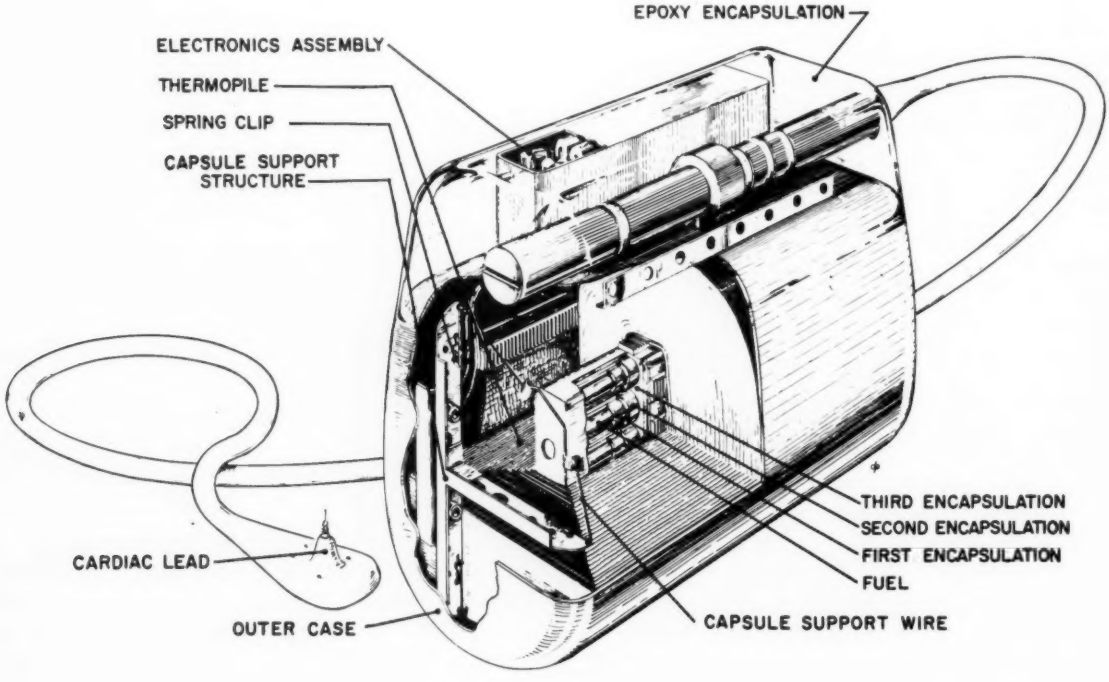

Fig. 6 USAEC–NUMEC isotopic power supply.

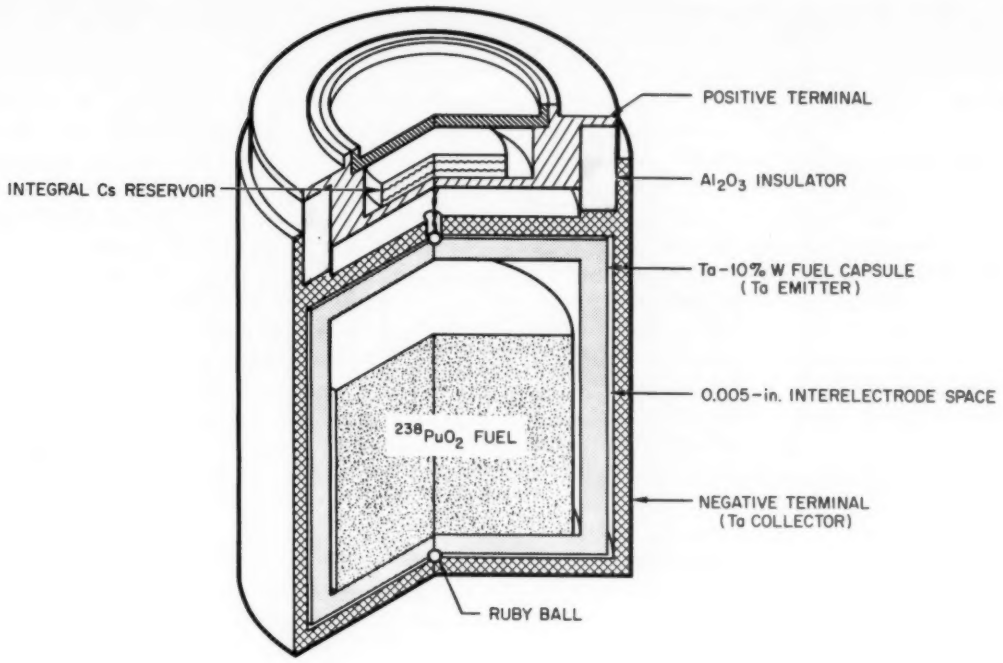

Fig. 7 Isomite power-source prototype fueled with ^{238}Pu.

and thus the emitter temperature. For example, a device of fixed size might supply electrical output ranging from 100 μW to 10 mW.

BETAVOLTAIC CONVERSION

The third energy-conversion technique is a nonthermal process. In betavoltaic energy conversion the decay heat from a beta-emitting fuel is converted directly to electrical energy by use of a semiconducting material that forms n–p junctions, which are coupled to planar sources of fuel material. Each beta particle striking the semiconductor, by means of ionization, causes approximately 10^4 electrons to be ripped from their bound states to produce free electrons. These electrons diffuse to the junction, where they are accelerated to the terminals by the junction electric field. The resulting electromotive force drives a current through the load.[10] A betavoltaic battery is constructed by encapsulating a number of these fuel and semiconductor wafers in metal and adding shielding and a ceramic insulation for the positive electrode.

Betavoltaic devices are practical only at power outputs of less than 1 mW. Therefore there has been less work done on them in the past than on thermoelectric or thermionic conversion devices. Betavoltaic devices have been built by various companies—e.g., Sandia Laboratories, Hittman Associates, and General Electric. Limited betavoltaic studies have also been done by the Australian Atomic Energy Commission. To the best of the author's knowledge, however, only Donald W. Douglas Laboratories is manufacturing betavoltaic devices today. Douglas' Betacel (Fig. 8) is fueled by ^{147}Pm, utilizes silicone semiconductor devices, and is encapsulated in stainless steel.

One advantage of a betavoltaic device is its versatility. For any particular application, the various characteristics, e.g., radiation level, volume, weight, and voltage, can be balanced by manipulating the fuel thickness, cross-sectional area, and number of wafers without changing the output power. As an example, a graph of ^{147}Pm layer thickness vs. maximum efficiency (Fig. 9) shows that an energy source with 10 pairs of wafers and a fuel thickness of 2 mg/cm^2 would have an efficiency of about 1.6%. A unit with a fuel thickness of 8 mg/cm^2 and only five pairs of wafers would have the same power output but an efficiency of only about 0.8%. The more efficient of the two systems would have a lower dose rate, but the less efficient would be smaller.[10]

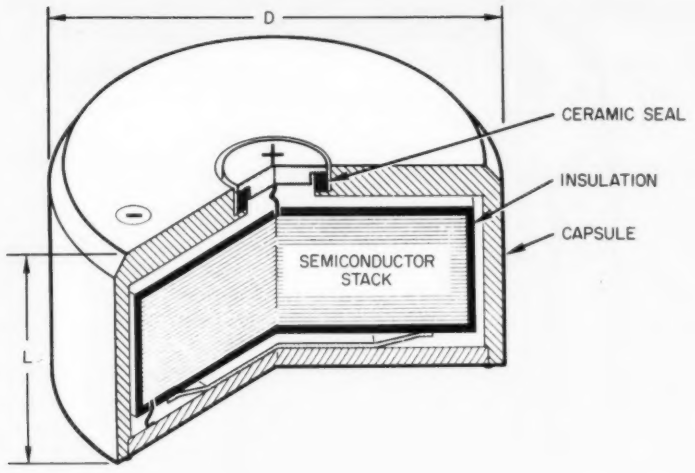

Fig. 8 Betacel—^{147}Pm fueled betavoltaic battery.

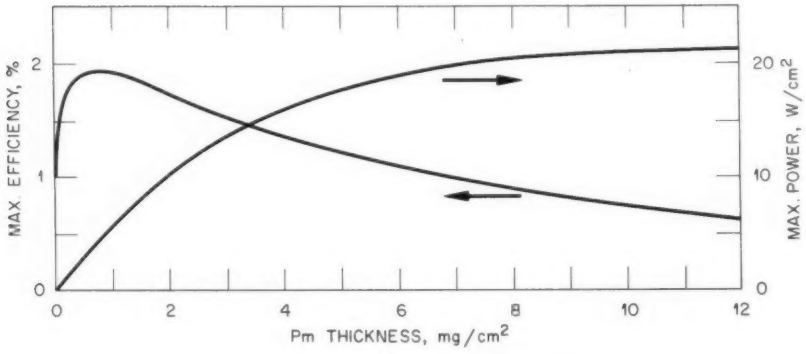

Fig. 9 Efficiency and power vs. ^{147}Pm fuel thickness for betavoltaic energy conversion.

COMPARISON OF FUELS

A very important aspect in the comparison of different generator types is the difference in fuel materials. The thermal methods of thermoelectric and thermionic conversion have primarily used ^{238}Pu—an alpha emitter of very high energy density and relatively low radiation dose rate when encapsulated. This dose is due to neutrons resulting from spontaneous fission of the plutonium and gamma photons from the daughter products of the ^{236}Pu impurity. The neutron dose rate in a given unit can be decreased only by decreasing the amount of fuel or by processing the fuel. The ^{236}Pu level—and consequently the gamma dose rate—can be decreased by neutron irradiation or aging.[11] In the United States, two purities of ^{238}Pu are readily available, one 80% pure ^{238}Pu with a ^{236}Pu level of ~2 ppM and the other 90% pure ^{238}Pu with a ^{236}Pu level of 0.25 ppM or less. These materials are available at $750 and $1250 per gram, respectively. The ^{238}Pu now available in France is about 85% pure and has a ^{236}Pu level of ~0.6 ppM. The ^{238}Pu expected to be available in Great Britain[2] will have a ^{236}Pu level between 0.5 and 1.5 ppM.

Nearly all betavoltaic devices have used ^{147}Pm, which has a high energy density and a beta energy ideally suited for this application. However, its 2.6-year half-life limits applications to periods of 5 to 7 years.

Further, the gamma radiation at the surface of the ^{147}Pm fuel capsule comes from the ^{146}Pm impurity, which can be decreased[12] by a factor of 2 or 3 by reirradiation in a reactor under carefully controlled conditions. The USAEC has not announced any plans to reirradiate a large quantity of ^{147}Pm, but it is understood that such is under consideration.

Other beta-emitting fuels that might be applied to betavoltaic conversion are ^{3}H and ^{85}Kr, with half-lives of 12 and 10 years, respectively. These half-lives would be ideal for isotopic pacemakers, and these isotopes also offer very low radiation levels when encapsulated. The chief disadvantages are the low energy densities, compared to ^{147}Pm, and the unattractive beta-particle energies—tritium beta energy is too low for efficient conversion and ^{85}Kr beta energy is high enough to cause semiconductor damage.

OVERALL COMPARISON

The most often compared parameter of the different radioisotope-fueled generator designs is energy-conversion efficiency. Efficiency is important, but, as fuel loadings are decreased to the point where the material cost is only a small fraction of the fuel encapsulation cost and acceptable radiation levels are obtained, other parameters, e.g., reliability, producibility, and cost, will become more important. In the future, for example, thermoelectric designs may become the most widely used because of the reliability statistics and experience gained in larger scale applications. On the other hand, because thermoelectric principles are highly developed, perhaps only limited technological advances will be made in the future. Because of the large number of thermoelements and their interconnections, thermoelectric devices might have inherently higher production costs and lower reliability than other systems.

Thermionic systems have a simpler design than thermoelectric devices and could potentially offer an advantage in reliability. Because of the close tolerances necessary between the emitter and collector, however, production rates might be lower and costs higher than for other systems. The question of whether a 200-μW thermionic device can attain efficiencies comparable to other systems is still unanswered, and doubts concerning the market potential may limit work on these smaller designs. Thermionic conversion is relatively new, however, and perhaps with time this technique will prove more applicable than the thermoelectric.

Betavoltaic devices seem to have the simplest design and construction combined with a high conversion efficiency and high power density. These factors, together with the low cost of ^{147}Pm ($0.20 per curie), suggest that the betavoltaic device may be the best of those discussed here for the implantable cardiac pacemaker.

The only difference between radioisotopic pacemakers and other types that have been used over the last 10 years is the radioactive material in the radioisotopic devices and the resulting radiation at their surfaces. In all radioisotope-fueled units designed for pacemakers with which the author is familiar, safe containment of the radioactive material has been the prime consideration. In the future, before its use in any prosthetic device, the fuel capsule will have to be qualified for safe containment during credible accidents, such as crush, impact, cremation, and gunshot. Many of the fuel containers discussed here have already passed these safety tests.

There has always been a question of what maximum radiation level is acceptable at the surface of the radioisotopic pacemaker. There do not seem to be any data that indicate a maximum tolerable radiation level for this application. Whatever the absolute value used in discussing radiation dose rate, two additional factors should be considered. First, for this application, the rem (roentgen equivalent man) should be used and not the radiation absorbed dose (rad), since here we are concerned with the actual effect of these radiations on human tissue. Thus for the ^{238}Pu-fueled devices, the radiation dose will be due not only to gammas (RBE = 1) but also to neutrons (RBE = 7 to 10). Second, rather than a maximum dose rate, an integrated average dose over the pacemaker life should be specified. This is important because different fuel materials are used. The dose rate from the ^{238}Pu-fueled devices increases with time as a function of the amount of ^{236}Pu present. The ^{147}Pm-fueled units show a decreasing dose rate with time.

With this consideration as a basis for comparison, the approximate average dose rates for the power supplies discussed above may be considered. The USAEC–NUMEC device now has[13] a beginning-of-life surface radiation level of 9.5 to 11.5 mrems/hr. The plutonium used is very "clean," and its average dose over 10 years should be only 11 to 12 mrems/hr. The British design reportedly will show an average 10-year dose rate of 9 mrems/hr. The French Alcatel unit has used a fuel containing "several" parts per million ^{236}Pu and shows a beginning-of-life dose rate[14] of 3 mrems/hr, which would average about 9 mrems/hr over 10 years. Currently available ^{238}Pu in France has 0.6 ppM ^{236}Pu. The 10-year averaged dose from

this clean French plutonium would be 6 to 7 mrems/hr. The ^{147}Pm-fueled units being designed for pacemakers will have a beginning-of-life radiation level of 14 mrems/hr, averaging about 9 mrems/hr over 5 years of operation.

Perhaps the biggest problem facing the isotopic pacemaker in the next year or two is not technical or economic but regulatory. Current government regulations for radioisotopes in the body were written for diagnostic testing, which utilizes radiochemicals in the bloodstream. When these regulations were set up, the concept of a qualified sealed source of radioactive material inside the body could not have been foreseen since it is a very new one. The USAEC, the ENEA (European Nuclear Energy Agency), and the UKAEA are studying possible regulation changes that would encompass this type of application. The timing and the final form of these regulations might to a large extent determine the availability of radioisotopic pacemakers in the future. (MG)

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Safety Features of French Cardiac Pacemaker Fueled with ^{238}Pu*

By R. Berger, R. Boucher, B. Jampsin, and M. Hot

Supplementary Keywords: *heart; prosthetic device; energy source.*

Abstract: *The properties of a ^{238}Pu source and the French (Alcatel–Medtronic) pacemaker are described.*

*Somewhat abbreviated version of a paper presented at the ENEA/Euratom Seminar on Radiation Protection Problems Relating to Transuranium Elements, Karlsruhe, Germany, September 1970, as printed in *CEA–ENEA Information Bulletin on Isotopic Generators*, **10**: 3-21 (November 1970).

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Isotopic energy sources are being introduced for powering cardiac pacemakers in order to increase the safety and the useful lifetimes of these devices. The mercury cells used at present are involved in most pacemaker failures, which are due to unforeseeable premature cell exhaustion. The interval at which

pacemakers need to be replaced—averaging 2 years with mercury cells—may be increased to at least 10 years by using isotopic power sources. On the other hand, the use of a radioisotope, e.g., ^{238}Pu, introduces the risks of irradiating the patient and his surroundings and possibly of contaminating the environment if the capsule should be ruptured.

This article discusses the use of ^{238}Pu in cardiac pacemakers with regard to pacemaker construction requirements, radiation safety of the patient, and integrity of the source capsule. The specific source on which the discussion is based was designed and constructed in France as a joint project of the Commissariat à l'Energie Atomique (CEA), the Société Alcatel, and the Broussais Hospital under the auspices of the Delegation Générale à la Recherche Scientifique et Technique. The safety criterions for the capsules were drawn up by the Study Group on Isotopic Batteries of the European Nuclear Energy Agency (ENEA).[1]

CONSTRUCTION REQUIREMENTS

Since cardiac pacemakers are implanted in the body, they must be as small as possible, and, in particular, their thickness is critical. Their density must be as close as possible to that of the neighboring tissues in order to avoid constraints due to gravity, and surgeons insist on rounded edges to simplify implantation. The surface temperature of the pacemaker must be as near body temperature as possible.

Pacemaking pulses are usually square waves, lasting from 0.7 to 2 msec, with a frequency of 55 to 80 per minute at a voltage of 3.5 to 7 V and a current of 3 to 15 mA. The minimum power consumption varies from 10 to 20 μJ per impulse, depending on whether the electrode is myocardiac or endocardiac. Precise specification of the minimum power needed is difficult, but a supply of 50 μW(e) to an asynchronous cardiac pacemaker and of 240 μW(e) to a relay pacemaker appears possible. Of the isotopic energy systems available, the best choice at present appears to be thermoelectric generators using Bi–Te thermocouples and a ^{238}Pu heat source. Interdependent factors affecting construction are thermal power required, size, weight, heat removal, and construction cost.

For fixed values of other source parameters, the thermal power required will decrease with decreasing dimensions of the ^{238}Pu capsule and of the radius and length of the source itself. Our research has shown that an energy source containing 150 mg of ^{238}Pu in a capsule 9 mm in diameter and 10.5 mm long, suitable for incorporation in a pacemaker casing 27 mm thick and 71 mm in diameter, can supply an electric power of 240 μW.

The materials making up the pacemaker, apart from the electronic components and the parts of the energy system (not counting the radioisotopic source and the thermoelements) must be as light as possible so that the pacemaker may have the same density as the surrounding tissues. Titanium is preferable to stainless steel, and conventional coating materials are replaced by an expanded resin. Thus, since the capsule provides the only effective shielding against gamma radiation, a thick cladding material of high density must be used. With a double cladding consisting of 1 mm of tantalum and 1.25 mm of iridized platinum, the capsule weighs 10.615 g. Since the average specific-heat production in human tissues is about 1.33 mW/cm^3, the heat supplied by 150 mg of ^{238}Pu can be dissipated in the body by conduction and convection in the blood without chronic irritation if the pacemaker volume is 65 cm^3. The pacemaker in question has a volume of about 100 cm^3 and thus provides a large margin of safety. The temperature gradient between the surface of the pacemaker and that of the neighboring tissue has been measured and found to be less than 0.1°C. Of course, any decrease in the required thermal power results in a saving in radioisotope and in shielding material.

SAFETY REQUIREMENTS

In the general use of cardiac pacemakers powered by isotopic energy sources, one must consider the possible risks run by the bearers and by the general population during normal operation of the devices and the possible consequences of foreseeable accidents that might lead to rupture of the capsule and perhaps to contamination. The design and test criterions for the capsules must take into account the fitting of the source into a cardiac pacemaker and the implantation of the device while it is operating, the likelihood of accidents, and the keeping of a permanent record of the devices since the recovery of all capsules may not be feasible. As far as possible, the safety of the energy systems should be such as to require a minimum amount of checking on the bearers of the pacemakers.

Radiation

The use of a radioisotopic source in a prosthetic device can be justified only if the medical advantage to the patient offsets the radiological disadvantage. Since

ionizing radiations damage living cells and an implanted pacemaker is in intimate contact with living tissue, the equivalent dose rate at the surface of the device must be as low as possible. The maximum annual permissible equivalent dose (30 rems/year to skin and bone) recommended by the International Commission on Radiological Protection (ICRP),[3] and applied in France[4] for persons who are occupationally exposed to radiation, could be used as a preliminary guide. This dose is conservative—there is no difficulty with equivalent dose rates of 100 mrems/day to the whole body—and is a feasible limit for construction purposes. Ultimately the permissible value depends on medical advice.

Radiation is sharply attenuated by tissues—by a factor of 10 in the first 2.8 cm of tissue (Fig. 1).

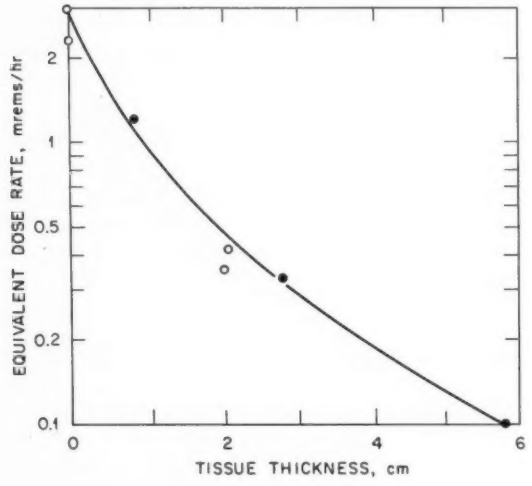

Fig. 1 Experimental (○) and calculated (●) equivalent dose rates from a 150-mg ^{238}Pu energy source in a cardiac pacemaker as a function of tissue thickness.

Fragile organs, e.g., the gonads and the spinal cord, and the whole body of the bearer will receive a very low equivalent dose, certainly less than 5 rems/year. The equivalent dose received by other persons, including those who live with the bearer, will be less than the ICRP limit of 0.5 rem/year for the whole-body dose to the general public.

The equivalent dose rates from the French source were measured by thermoluminescent dosimeters (Teflon loaded with LiF) for gamma radiation and by nuclear emulsions for neutrons, both of which are conventional techniques. An assembly consisting of three thermoluminescent detectors and a nuclear emulsion was used for measurements on the skin. Values for the equivalent dose due to neutrons are based on the quality factors adopted under French law.[5]

The results (Table 1) show an average measured dose rate in contact with the pacemaker of about 2 mrems/hr for gamma radiation and about 1 mrem/hr for neutrons—a total of 3 mrems/hr at the "hottest" point. The average gamma-radiation reading on the skin of the bearers at a point as near as possible to the source (about 2 cm away) was 0.25 mrem/hr, and the neutron dose was too low to be measured accurately. These few experimental values agreed with calculated values (Fig. 1).

The equivalent dose rates from the pacemakers depend on the purity of the ^{238}Pu used and on its chemical state. The chief troublesome impurities are ^{236}Pu and the light elements Be, B, F, Li, Na, Mg, Al, C, Si, and O. The gamma contribution from ^{212}Pb, ^{212}Bi, and ^{208}Tl—daughters of ^{236}Pu (at an atomic

Table 1 Maximum Equivalent Dose Rates from Implanted Pacemakers

Exposure time, hr	Tissue thickness, mm	Equivalent dose rate, mrems/hr		
		γ	n	Total
95	0	1.5	0.8	2.3
360	~10	0.5	0.18	0.68
311	~10	0.4	0.11	0.51
117	0	2.0 3.1 1.7	1.1 0.85	3.2
~200	0	1.92 1.95	1.0 1.3	3.1
451	20	0.28 0.20 0.26	<0.09	<0.34
138	0	2.3	0.43	2.7
284	21	0.23 0.22 0.27	0.17	0.41
~200	0	1.66 1.69	1.2	2.9
140	0	1.77	1.6	3.4
783	21	0.38 0.42 0.49	0.05	0.48
307	0	1.95	0.8	2.75

concentration of 10^{-6})—equals that from initially pure ^{238}Pu after 3.5 years (Fig. 2), and hence an average dose over the whole mission life of the pacemaker must be calculated. In practice, a ^{236}Pu concentration of about 0.3 atom per 10^6 atoms of ^{238}Pu can be obtained economically. Fast neutrons resulting from nuclear interactions between the alpha particles emitted by the ^{238}Pu and the light elements mentioned above may also be present. The use of ^{238}Pu

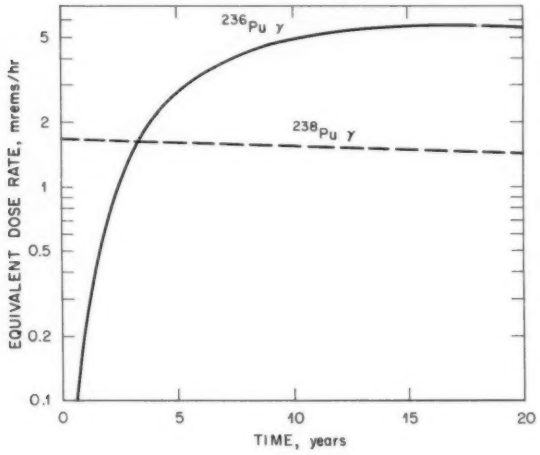

Fig. 2 Time variation of equivalent gamma dose rate from a 150-mg ^{238}Pu energy source at contact with the outer surface of a cardiac pacemaker at the midplane of the pellet (1 atom of ^{236}Pu per 10^6 atoms of ^{238}Pu).

metal rather than oxide avoids the presence of large amounts of oxygen.

Calculated[6] equivalent gamma dose rates at points on straight lines parallel to the source axis, in the plane containing this axis but normal to the planes of the ends of the pacemaker (Fig. 3), showed that the gamma emission from isotopes heavier than plutonium is negligible compared with that from ^{238}Pu. The same applies for ^{238}Pu fission gammas and fission products. The rates were obtained with the Mercury 3 code[7] in which propagation of the particles was calculated from a linear attenuation model with an accumulation factor, using the CEA library of effective gamma cross sections and accumulation factors.[8]

The ANISN code[9] was used to calculate the equivalent neutron dose rates. This program solves the one-dimension Boltzmann equation by the method of discrete ordinates in neutron transport and treats anisotropic diffusion of any order. The calculation uses

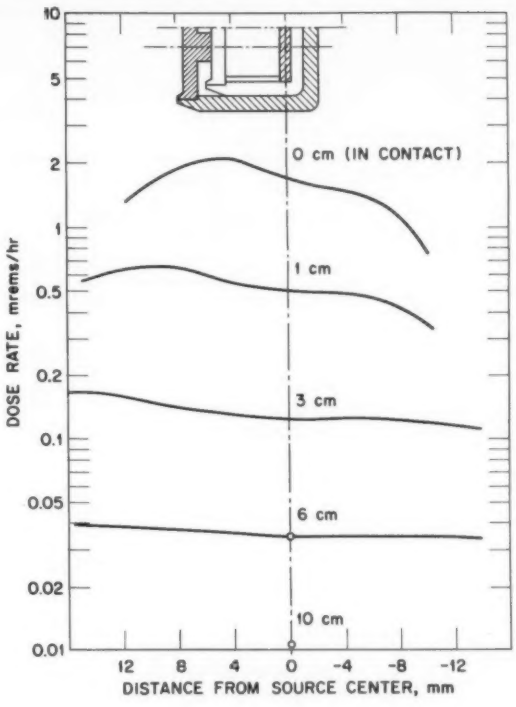

Fig. 3 Change in equivalent gamma dose rate from a 150-mg ^{238}Pu energy source in a cardiac pacemaker as a function of tissue thickness.

99 groups. The multigroup neutron library used is derived from the ENDF/B valuation.[10] Only the neutrons emitted in the spontaneous fission of ^{238}Pu were considered.

Thermomechanical Tests

The radioisotope is clad in a sealed, leakproof capsule. Containment must be ensured under normal operating conditions and in foreseeable accidents during the whole mission life plus a sufficient margin of time to cover contingencies, and account must be taken of the increasing internal pressure due to the accumulation of helium in the capsule. Calculations on the development of the helium pressure and of the temperature of a capsule containing 155 mg of ^{238}Pu for an initial temperature of 70°C (Fig. 4*a*) show that after 1000 years the internal pressure reaches 360 atm and the temperature, 20°C. After 100 years at 1000°C, internal pressure reaches 830 atm (Fig. 4*b*).

The latest edition of *Vital Statistics of the United States* (1967) shows that the most likely fatal accidents

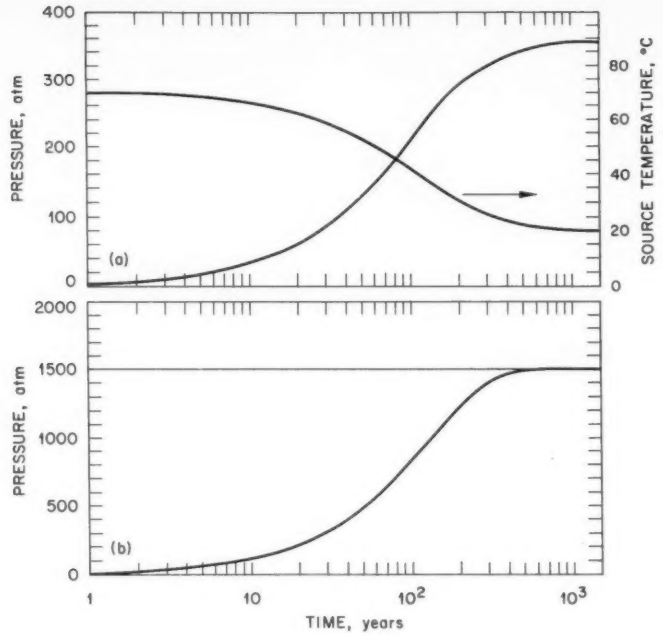

Fig. 4 Thousand-year values for (*a*) variation of pressure and temperature and (*b*) pressure at 1000°C for a 155-mg ^{238}Pu energy source.

are due to motor vehicles, firearms, fire and explosion of fuels, cutting and penetrating instruments, machines, and aircraft. Such data for Europe are different as regards firearms because of European firearm laws. The CEA (CEN at Fontenay-aux-Roses) has made all tests required by the existing regulations[11,12] regarding transport in order to obtain acceptance of the French sources as "special design." These tests (Table 2) have shown these sources to be resistant to forces of any expected accident.

Percussion Test. In the United States a careful study has been made[13] on the probability that a bullet might destroy the capsule of a radioisotopic pacemaker. Taking into account the cross-sectional area of the capsules (about one one-thousandth that of the vital organs), the average age of pacemaker bearers (about 70 years, which is high compared with that of persons killed by firearms), and the type of firearms and bullets used in different circumstances (suicides, accidents, or murders), the probability of rupture of a pacemaker capsule by a bullet was found to be 6.3×10^{-5}, which is equivalent to 6.3 accidents in 100 thousand years for a constant population of 20 thousand bearers. This probability may be decreased to 3.8×10^{-6} by keeping the bearers away from lethal firearms, and to 1×10^{-6} if the containment of the radioisotope is required to remain intact only under the direct impact of the maximum charge of hand guns and shotguns as well as of rimfire .22 caliber bullets. (Probabilities are currently less than 1×10^{-6} in Europe and Japan.) In the event of the cladding's being ruptured by high-powered-rifle bullets, the plutonium released would probably be limited to the body of the bearer and the blood from the wound.

Percussion tests on the French capsules are now in progress in western Germany (Physikalische-Technische Bundesanstalt, Braunschweig, Bayerisches Landeskriminalamt, and Technische Universität, Munich), but the U. S. conclusions clearly show that no capsule of a size compatible with surgical implantation could resist the impact of projectiles from all firearms that can be purchased.

Crush Test. The probability that a capsule will be compressed by the collapse of a building or by the passage of a rigid wheel of a vehicle such as a railway wagon is extremely low, although the load applied to the capsule in such cases would be very high, e.g., 1.7 to 10.7 tons for the wheel of a railway carriage. An overload of 30 to 40% must be added to these values to allow for the dynamic effect.

Table 2 Tests for Sealed Sources

Tests	Test temperature, °C	Internal pressure, kg/cm²	Nature of test	Effect on source
Shock resistance	100	30	Free fall through 9 m	Very slight deformation, no leakage
Percussion	100	30	Weight of 1.4 kg falling from 1 m onto the source	Very slight deformation, no leakage
Crushing	25	Vacuum	Load of 1500 kg	Very noticeable deformation, no leakage
Impact	100	30	Source speed of 50 m/sec	Two sources showed leakage from platinum cladding (but not from tantalum cladding); one suffered additional damage during test
Internal pressure resistance	1000	840 (810 atm)	Maintained 30 min	No dimensional change, no leakage
	1200	605 (580 atm)	Maintained 20 min	Slight deformation of cladding, no leakage
Fire resistance	1300	Vacuum	Maintained 6 hr in air	Liquid Pu alloy reacts with the Ta unless Ta is carburized; no damage to external iridized Pt cladding
	800, then 20	30	Maintained 15 min in air at 800°C, cooling to 20°C	No damage
Water immersion	30		*p*H = 7, 1 week	No damage

The most realistically foreseeable accident for the capsules is one due to compression by a pneumatic-tired wheel or to a frame collapsing. In the case of a truck, the pressure is estimated to be less than 10 kg/cm², whereas with a collapse the load can reach 1000 kg. It should be noted that the plutonium alloy with face-centered cubic structure is particularly malleable and may be deformed without cracking or crumbling.

The CEA has made a series of tests on about 10 capsules. Loads of 150 or 500 kg applied to the Pt–Ta capsules on one end or along the surface of the cylinder caused very little deformation; 1500 kg caused noticeable deformation but no rupture or leakage. Leaktightness was checked by the method of "helium sniffing," using a mass spectrometer calibrated with helium and having a sensitivity limit of 10^{-11} atm-cm³/sec. The results of tests carried out in Spain by INTA (National Institute for Aerospace Techniques) at 150 and 500 kg and 100°C, using an Instrom machine, on two tantalum capsules and two Pt–Ta capsules were identical to those of the CEA.

Impact Test. An aircraft accident is a foreseeable instance where the capsules acquire the highest speed with respect to fixed obstacles. The maximum speed has been estimated as 50 m/sec by comparison with the safety conditions required for flight recorders (100 to 1000 g during a period of 5×10^{-3} sec) and evaluating the limiting speed of the capsule in falling. In tests made by INTA, tantalum capsules with an impact speed of 50 m/sec on an 18-mm-thick steel plate normal to the direction of travel (internal capsule pressure 30 kg/cm² and temperature 100°C) did not leak. Of 12 Pt–Ta capsules similarly tested, two showed leakages—4×10^{-6} and 3×10^{-6} atm-cm³/sec, respectively—in the external platinum capsules, the second capsule having been damaged additionally by a collision with the arm of the apparatus.

These leakages were due to welding defects, and the internal tantalum claddings had kept their integrity. As forecast by the probability calculation concerning the position of the capsule on meeting the obstacle, all impact marks were on the directrixes in the end planes.

Thermal Test. From statistics of the National Fire Protection Association (NFPA), the United States has found a probability of 10^{-6} that a cardiac pacemaker would be exposed for 2.63 hr at a maximum temperature of 1038°C in an ordinary fire. Refuse fires last 15.33 hr at 538°C on an average, whereas cremations last from 1 to 2 hr, with an ideal temperature of 870°C, a minimum of 735°C, and peaks of up to 1370°C for 10 to 15 min. However, the probability that a capsule would be subjected to these two kinds of fire is very low. In fact, bodies caught by fire are usually on the ground, where temperatures are significantly lower than at higher levels; they are wetted during fire fighting; and they are usually recovered. The NFPA has never recorded a case of complete incineration. In 250 deaths by fire, only one case of incipient incineration has been found.

By comparison with the test for containers of nuclear materials involving exposure to fire for 30 min at 800°C, and taking account of the fact that the isotopic energy source is built into a pacemaker which is itself implanted in a human body, it is very unlikely that the temperature of the source would exceed 500°C in an ordinary fire. Seven Pt–Ta capsules maintained at 1000°C in air successively for 0.5, 2, and 6 hr showed no damage. The capsule materials have a high melting point—3000°C for tantalum and 1820°C for Pt–20 wt.% Ir alloy—and capsules maintained at 1000 and 1200°C for 30 and 20 min with internal pressures of 810 and 580 atm, respectively, showed a slight deformation of the cladding at 1200°C but no leakage.

Chemical Corrosion Tests

The capsule material must remain compatible with the fuel at its highest operating temperature, and the containment must not be compromised by chemical attack on the capsule material either in normal operation or in the event of any foreseeable accident.

Compatibility. Tantalum and iridized platinum have excellent compatibility with Pu–Sc alloy. The diffusion of liquid plutonium between the grains of the tantalum may be completely stopped by surface carburization of the internal wall of the cladding. The liquid plutonium forms, with the iridized platinum, metallic compounds of relatively high melting point, which act as a diffusion barrier. During 0.5- to 2-hr thermal tests at 1000°C, a significant reaction zone was observed when the plutonium alloy had been placed directly in a platinum capsule, but after that time there was no further development of the reaction zone. The metallic compounds were identified by micrography and analyzed with an electron probe.

Chemical Inertness. Chemical agents likely to come into contact with a capsule are air, fresh water or seawater in the event of drowning, and organic matter from the body and from the earth in the event of the patient's being buried without recovery of the pacemaker. The Pt–Ir cladding is particularly resistant to oxidation at high temperatures, and the Pu–Sc alloy showed no oxidation or variation in weight when kept 20 months in 30°C air at 60% relative humidity. Pure plutonium is oxidized after being kept 1 hr at 150°C, but Pu–Sc alloy undergoes successive thermal treatment going from 100 to 400°C over a period of about 200 hr before being oxidized.

The corrosion resistance of platinum in fresh water, seawater, saline solution, or organic matter is well known. (MG)

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Editor's Note: The overall French pacemaker program was described in *Nuclear News*, **13**(2): 42-46 (December 1970), based on a paper presented at the American Nuclear Society Meeting of Nov. 15–19, 1970, Washington, D. C.

SUBMINIATURE PACEMAKER TOTALLY INSIDE HEART

A tiny pacemaker (Fig. 1) has been developed which can be inserted entirely into a chamber of the heart by only minor surgery. The device was developed at the Cox Heart Institute of the Kettering Medical Center in Dayton, Ohio. The concept has been shown to be feasible in several animal implants, using devices powered by conventional chemical batteries or by a Betacel atomic battery provided by the McDonnell Douglas Corp. through its Donald W. Douglas Laboratories of Richland, Wash. This accomplishment, believed to be the first of its kind, was achieved entirely with private funds, including development of both the pacemaker and the atomic battery. It is the result of a joint effort by medical scientists and engineers led by Dr. J. W. Spickler, physiologist at the institute, and Dr. N. S. Rasor, a consulting physicist for the Douglas Laboratories and for the institute.

Space-age developments in solid-state electronics and atomic energy have permitted the pacemaker and power source to be greatly miniaturized. The experimental Cox device is a capsule less than $^{5}/_{16}$ in. in

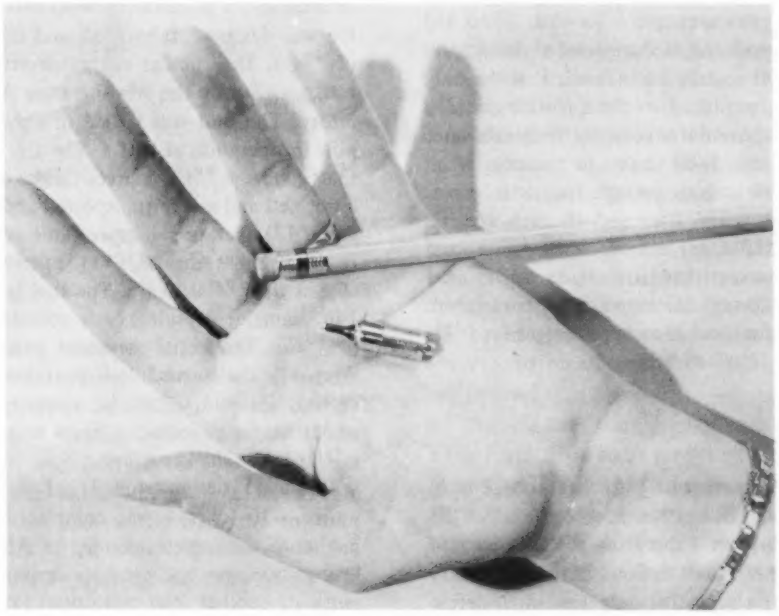

Fig. 1 Subminiature pacemaker.

diameter and ¾ in. long—about the size of a pencil eraser. It is small enough to be inserted into a vein in the neck by minor surgery and then passed into a chamber of the heart, where it is attached to the inner wall by a mechanism similar to a safety pin. The insertion device is then detached and removed, and the incision is permanently closed.

The device may be ready for clinical use in about 2 years.

Alternative forms of the atomic battery, which use longer life nuclear fuels, are being developed by the Douglas Laboratories. Such batteries can potentially operate the pacemaker capsule for several decades without replacement. Through this development it eventually should be possible for even a child to have a lifetime "built-in" artificial pacemaker implanted in its heart by minor surgery. (MG)

^{60}Co Use in Thermoelectric Generators

Summarized by Martha Gerrard

Supplementary Keywords: *energy source; thermoelectric system; conversion, electric.*

Abstract: *The advantages of ^{60}Co as a heat source for use in thermoelectric generators are discussed, and several conceptual and prototype designs are described.*

A number of properties of ^{60}Co make it of interest for use in heat and power sources.[1,2] Its cost, $7 to $15 per thermal watt produced, is the lowest of that of any of the radionuclides usually considered. It is the only isotope now being produced in the quantities needed to make large sources, and it is comparatively safe since it does not enter into food chains in the case of an accident. Its half-life is long enough for use in power systems of several years' life, and its high specific activity (200 to 450 Ci/g) can result in lighter and more compact sources than are made with other radioisotopes even though thicker shielding is required.

Several conceptual and prototype designs for ^{60}Co energy sources are described here.

U. S. STUDIES

The major development of ^{60}Co heat-source technology in the United States has been centered at the USAEC's Savannah River Laboratory (SRL), operated by E. I. du Pont de Nemours & Company, Inc. Primary emphasis has been on providing data needed to define allowable long-term operating conditions for ^{60}Co capsules and to evaluate the safety of ^{60}Co heat sources. With this aim, the compatibility of cobalt metal with superalloy capsules at 850 to 1000°C and with refractory or noble-metal capsules at 1000 to 1200°C and of cobalt compounds with refractory or noble-metal capsules at 1200 to 2000°C has been investigated.[1]

As a practical demonstration of the ^{60}Co fuel technology, a nominal 90-W(e) thermoelectric generator was designed, fabricated, and fueled with 183 kCi of ^{60}Co. The tubular thermoelectric module (Fig. 1) was provided by the Westinghouse Astronuclear Laboratory. The unit was fueled in February 1961 and is now in operation at SRL.[3] The test has demonstrated that: (1) large ^{60}Co sources can be easily assembled in a hot cell and safely transported and used in inhabited areas, (2) operating temperatures are low enough for practical long-term use, and (3) performance is predictable as the ^{60}Co decays. The fuel is in two 4-in.-long, 1-in.-diameter capsules, each containing about 90 kCi of ^{60}Co. The initial measured power output was 87 W(e). The fueled module is contained in a ductile iron biological shield, 39 in. in diameter and 48 in. high, which decreases radiation doses to approximately 200 mR/hr at the shield surface.

Detailed design (Fig. 2) of a large ^{60}Co heat source—30 kW(t)—was completed in 1970 at Westinghouse Astronuclear under an AEC contract.[4] The heat-source concept provides operability over a wide range of coolant inlet and outlet temperatures, which should make it usable with a variety of conversion systems and applicable for a dry terrestrial environ-

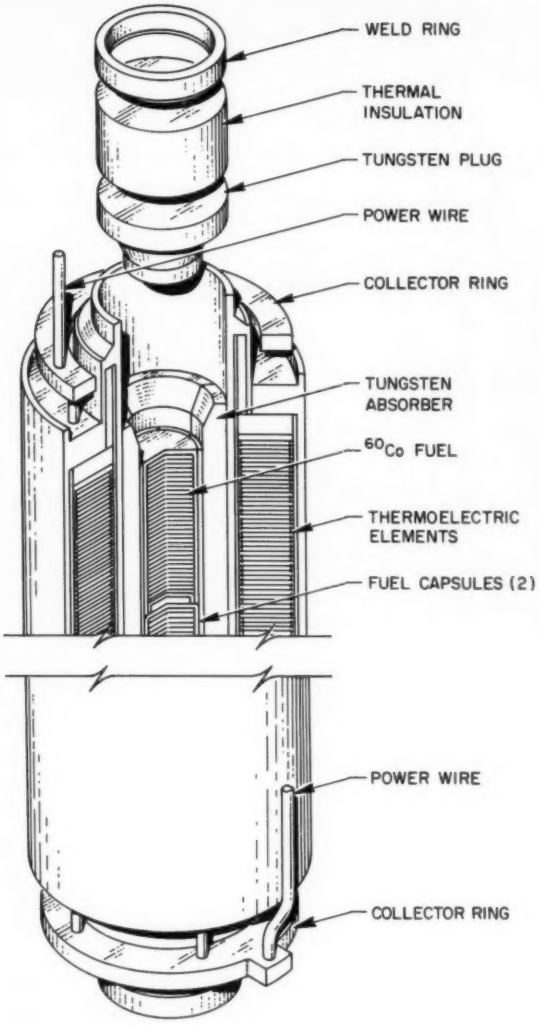

Fig. 1 Thermoelectric module for ^{60}Co test at SRL.

ment. The gas-cooled source is being fabricated and assembled, and demonstration testing is scheduled for 1972. Outlet temperature capability is up to 1350°F. A passive emergency cooling system consists of 36 sodium heat pipes, qualification tests for which are under way. The fuel-pin assembly provides double encapsulation of the fuel, which is in the form of standard SRL nickel-plated wafers. The cobalt wafers have been irradiated to specific activities up to 400 Ci/g.

Following a basic test program, plans call for mating of the source with existing dynamic power-conversion equipment—Brayton and organic Rankine—for demonstration testing of multi-kW(e) isotope power systems.

MAPLE GENERATORS

The Canadian MAPLE (*M*inor *A*tomic *P*rolonged *L*ife *E*quipment) power source was designed[5] for fueling with the 75- to 100-Ci/g ^{60}Co expected to be produced in the Canadian hydro reactors, such as those at Pickering. The MAPLE IA model—a laboratory prototype—was followed by the MAPLE IB, a production prototype that is now operating a navigation light on the St. Lawrence Seaway. Another MAPLE I unit, using uranium shielding, is being developed to provide heat and electric power for an Arctic weather-data-collection system.

The MAPLE IB (Fig. 3) unit, described here, was charged with 15.7 kCi of ^{60}Co on Sept. 14, 1970, and is expected to have a useful life of 5 years. Its heat output is 43 W(t), and its total weight is 5500 lb. Its transport container classification is International Class B.

The source is made up of seven ^{60}Co pencils, each with an inner capsule of 316L stainless steel and an outer one of Deloro C (Deloro Stellite equivalent of Hastelloy alloy C). The pencils are surrounded by a U–8 wt.% Mo alloy heat block, which is suspended from the copper closure plate by three heat-dump tubes (source-temperature-stabilizing units) clamped to three of the six heat-block surface flats. Thermoelectric modules (Bi–Te alloy) are clamped to the heat block, and the whole thermoelement region is enclosed in a sealed jacket, which is filled with inert gas. Voids around the modules are packed with a microporous insulation. Following tests on the unit to determine its response to environmental temperatures from +30°C to −30°C and to demonstrate its compatibility with the navigation-aid system, it was installed in a navigation-aid tower on the government wharf at Brockville, Ontario, to provide power for a St. Lawrence Seaway navigation light.

The module configuration used allows power of 1 W(e) and more to be generated at on-load voltages of more than 1 V. The basic array is a 10 by 10 matrix of alternate *p*- and *n*-type Bi–Te alloy limbs. These matrixes, made of polyimide materials, have high radiation and temperature stability.

Because of the short (5.26-year) half-life of ^{60}Co, stabilization of the source temperature with declining thermal power is required. Mercury reflux tubes were

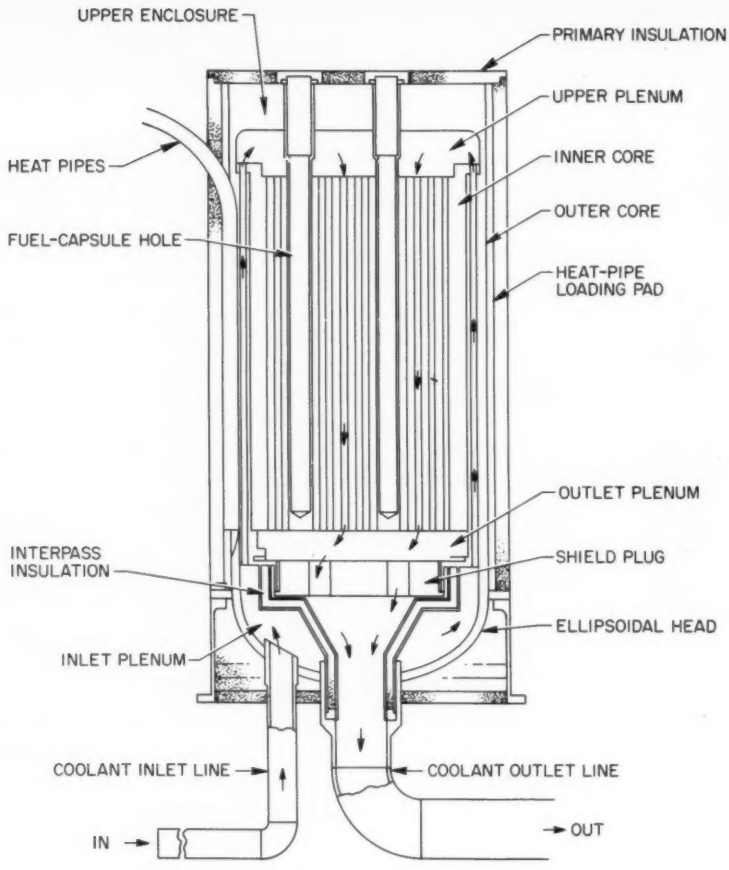

Fig. 2 Westinghouse ^{60}Co heat-source core assembly.

selected for this purpose, chiefly because mercury is stable to radiation, is thermally stable up to 300°C, has a vapor pressure <1 atm at 300°C, and has good heat-transfer properties. Tantalum, despite its cost, was selected for the tube material because of its outstanding resistance to attack by mercury. These tantalum tubes have a copper sleeve brazed to the top outer section to improve heat transfer and are filled with helium gas. Many more tubes are provided than the minimum needed. In the first few years of operation, when excess power is available, the electric-power output rises considerably as the environmental temperature drops, making the unit ideal for locations with high winter demands.

A simple two-transistor Royer-type circuit is used to step the generator output voltage up from approximately 4.5 to 13 to 15 V dc for charging a set of sealed Ni–Cd batteries. Silicon transistor units were selected because the initial efficiency of 80 ± 1% had not decreased after 15 thousand hours. Sintered-plate batteries were used as an intermediate stage for navigation-light systems to allow storage of power generated during the day.

MAPLE IB has been licensed by the Canadian Atomic Energy Control Board for (1) transport by surface routes in accordance with the standards of the IAEA Safety Series No. 6, Regulations for the Safe Transport of Radioactive Materials, 1967 edition, and (2) use in a public area of a well-populated region in Canada.

GICODYNE GENERATOR

The GICODYNE (*G*enerator, *I*sotopic, with *CO*balt using thermo*DYN*amic conversion), a 400-W(e) radioisotopic generator, went into experimental operation

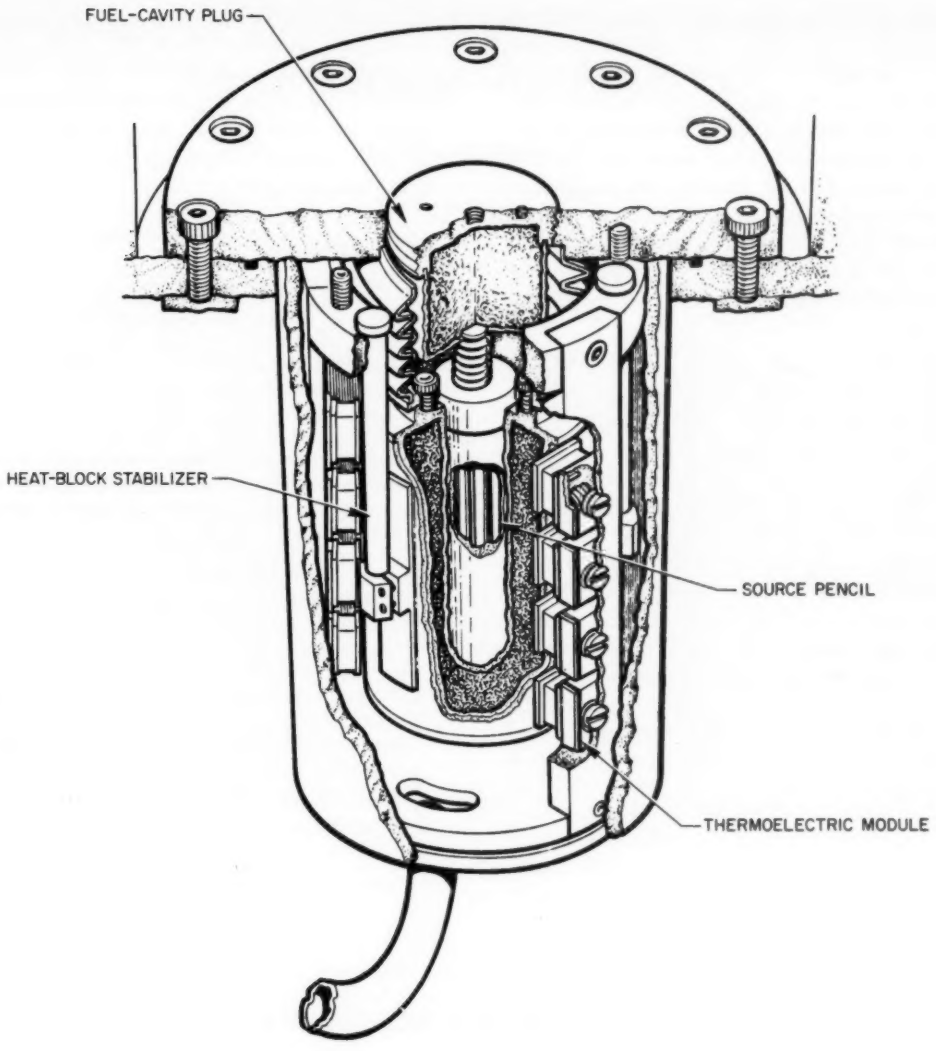

Fig. 3 MAPLE IB core assembly.

Mar. 3, 1971, at Serans in the Oise Province, near Lierville, France.[6] The purpose of this prototype unit is to demonstrate the feasibility of supplying a Hertzian relay for transmitting color television between Serans and Levallois. It is expected to operate for 2 to 2.5 years without external intervention. It may also be used to heat underwater structures and to supply lighthouses or signal buoys, radio navigation beams, and underwater petroleum wellhead controls.

The assembly weighs 4500 kg and is 5.22 m high and 0.9 m in diameter. The heat source, which is made up of 580 plaques of ^{60}Co—310 kCi initially—and is encapsulated in stainless steel, releases about 4450 W(t). The cobalt is kept in a helium atmosphere to ensure thermal homogeneity. The conversion system is a closed-circuit Rankine cycle with an organic liquid as the fluid. The overall efficiency of the system will be relatively low—7%. However, research is continuing at the Nuclear Studies Center of Grenoble, especially on the turboalternators, to achieve an efficiency of 20%; an efficiency of 12% would be considered satisfactory at present.

The radiation level at the outside is always less than 200 mrems/hr at wall contact. This tight heat source is immersed in the liquid to be vaporized, which is contained in a stainless-steel calandria-type evaporator that is externally heat insulated by a lamination of low-conductivity layers and reflecting layers. The calandria assembly is installed at the use site, and its junction with the energy-conversion union is made with a joint that is fusible at 320°C. In case of breakdown, the calandria would be released at this joint, and the heat source would be laid bare and would cool by natural-air convection.

Of the heat released by the cobalt, 95% is transmitted into the copper blocks and 5% into the laterally positioned uranium. The Gilotherm DO (diphenyl–diphenyl oxide) liquid in which the source is immersed is vaporized and condenses on the calandria plug, where it transmits its heat to the converter fluid. The converter fluid is also Gilotherm DO plus dichloro- or monochlorobenzene. These fluids do not undergo radiolysis or pyrolysis, have relatively low boiling temperatures, and provide self-lubrication of the turbine gas bearings. The two stages of the converter are hermetically sealed, each with a turbo-alternator. The cover of the calandria is the bottom of the boiler of the first converter stage. The second stage is heated by the vapor from the first-stage turbine and is cooled naturally by ambient air.

The GICODYNE is the first isotopic generator constructed in France to supply electric energy by thermodynamic conversion in closed circuit. This technique will allow making autonomous electric generators of from several tens of watts to several kilowatts, powers that enlarge the field of utilization of isotopic generators.

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ISOTOPIC ELECTRIC GENERATORS

By F. Laveissiere, French Report CEA-BIB-190, January 1971. (English translation: USAEC Report ORNL-tr-2485 by Martha Gerrard.)

Supplementary Keywords: *energy source; bibliography.*

This extensive listing of radioisotopic energy sources, their characteristics, the isotope used, and the literature sources of the information given will be useful to persons in this field. The inevitable typographical errors in a compendium of this length are unfortunate, but reference to the original source can be made when data are questionable. The isotopes concerned include ^{90}Sr, ^{238}Pu, ^{210}Po, ^{147}Pm, ^{242}Cm, ^{144}Ce, ^{170}Tm, ^{137}Cs, ^{227}Ac, ^{60}Co, and mixed fission products.

Comparative Evaluation of Power-System Types for Underseas Applications

By Robert E. Wimmer*

Supplementary Keywords: *energy source; comparison; sea.*

Abstract: *Comparison of possible energy sources for underseas missions indicates that as the mission time duration increases, batteries give place to fuel cells and they, in turn, to nuclear systems. Isotopic systems are preferable to nuclear reactor systems only at low power levels.*

Various power sources for underseas application are available or are being developed and tested. The purpose of this article is to identify key parameters of likely power systems and of missions and to suggest methods and criterions for evaluation in the overall mission context. Power systems considered are batteries, fuel cells, and both radioisotopic and nuclear-reactor thermoelectric and dynamic systems.

POWER-SYSTEM PARAMETERS

In underseas mission planning, the weight of the power system has been overemphasized—few power-system packages will sink unless ballasted and frequently volume and density are more significant than weight. In some missions, particularly manned stations, by-product heat may be either a benefit or a detriment, depending on mission conditions. And, of course, costs of both initial investment and support are always important.

The parameters usually examined in an analysis of a nonpropulsive power plant for underseas missions are size, cost, fuel consumption, density, efficiency, and life, which are compared in Table 1 for battery, fuel-cell, radioisotopic, and nuclear-reactor systems. For both batteries and fuel cells, all parameters vary linearly over the wide power range of interest—watts to kilowatts. This is not true of radioisotopic and nuclear-reactor devices.

All the batteries considered are primary, although Ag–Zn batteries (and modern versions of the conventional lead–acid battery) are attractive as a secondary source in some cases, e.g., propulsion. In general, however, it is unlikely that these batteries will become widely used. The chief merit of the laminated Leclanche battery is its low cost, but currents are limited to a few milliamperes per cell. Battery performance may be less satisfactory at temperatures outside the 70 to 80°C for which the ratings are given here.

With regard to fuel cells, the Hydrox cells are not a specific hardware item but rather are representative of hydrogen–oxygen cells generally. The $Zn–O_2$ cells, although usually referred to as "batteries," are classed here as fuel cells because of the nature of their operation (mechanical recharge).

The radioisotopic-power-system parameters cited are based principally on a study of ^{60}Co devices in the multikilowatt region. Figure 1 shows the dependence of cost and size on power level. Extension of the curves to 20 or 30 kW(e) (Fig. 1*b*) is not intended to imply that such power levels are currently deemed feasible, since no attention was given to practical problems such as source handling and system integration. These latter considerations also appear to limit active interest in isotopic systems to power levels below about 10 kW(e). The few data points in the low-power region lend credence to the extrapolation. Flattening of the RTG (*R*adioisotope *T*hermoelectric *G*enerator) specific cost curve in the region above 1 kW(e) is due to the increasing dominance of isotopic-source and -converter costs, although this situation may change as more fully evolved isotopic-source-packaging cost assessments are developed. Weights include shielding for onshore handling. There is little dependence of weight and volume on the type of power conversion used, and the radioisotope-system density—about 300 lb/ft^3—is nearly independent of system power level because of the large fraction of system weight allotted to dense shielding.

The costs, weights, and volumes shown for nuclear-reactor power plants in Fig. 2 do not represent any specific design but are a composite representing present and anticipated near-future developments. Various power-conversion systems can be used. Compact, liquid-metal-cooled reactors of the SNAP type

*Formerly at North American Rockwell Corporation, Atomics International Division; now at Litton Systems, Inc., Van Nuys, Calif. This is a shortened version of a paper presented at the 6th Annual Marine Technology Society Marine Technology Conference, Washington, D. C., June 29–July 1, 1970, Preprints Vol. 2, pp. 1289-1299.

Table 1 Nonpropulsive-Power-Plant Parameters*

Power-plant type	Unit weight, lb/kW†	Unit volume, ft³/kW	Unit cost, $/kW	Fuel consumption			Density, lb/ft³	Thermal efficiency, %	Nominal life, hr
				lb/kW-hr	ft³/kW-hr	$/kW-hr			
Battery (70 to 80°C)									
Leclanche laminated	‡	‡	‡	20	0.23	6	87	90	~2,000
Alkaline	‡	‡	‡	21	0.18	21	117	90	~5,000
Mercury	‡	‡	‡	18	0.15	90	120	90	>5,000
Ag–Zn	‡	‡	‡	12.5	0.10	150	125	90	10,000
Fuel cell									
Hydrox,									
pressure storage	100	3	10,000	19	0.32	0.10	59§	60	3,000
cryo storage	100	3	10,000	0.94	0.032	0.10	29§	60	3,000
$Zn–O_2$,									
pressure storage	450	9.4	10,000	7.0	0.096	15–20	73§	80	40,000
cryo storage	450	9.4	10,000	3.1	0.032	15–20	97§	80	40,000
Radioisotopic									
Thermoelectric	540–1,100	1.8–3.6	215,000–4,000,000	¶	¶	¶	300	5–10	40,000
Dynamic	540–1,100	1.8–3.6	50,000–150,000	¶	¶	¶	300	10–20	10,000–20,000
Nuclear reactor									
Thermoelectric	700–10,000	35–600	20,000–70,000	¶	¶	¶	20–30	5–10	20,000–80,000
Dynamic	16–4,000	0.5–200	700–20,000	¶	¶	¶	20–30	10–30	10,000–20,000

*Data for 1969.
†All power levels are kW(e).
‡Not applicable since batteries are not limited by power-density considerations.
§Fuel only, packaged.
¶Not applicable since nuclear fuels are considered a part of the power plant.

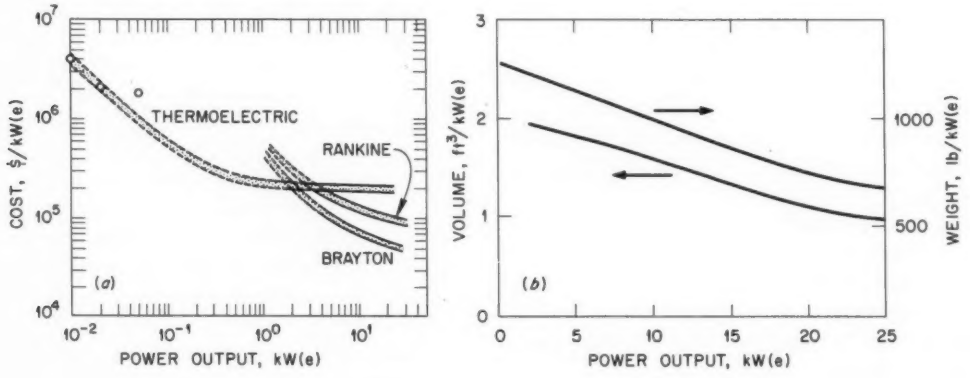

Fig. 1 (*a*) Cost and (*b*) size of radioisotopic power plants as a function of power output.

have been extensively proof-tested, and are directly applicable in the lower power region. At levels greater than several hundred kW(e) (per single unit), the larger conventional water reactors become more attractive. Weights given in Fig. 2 include sufficient radiation shielding for postoperational handling and prevention of neutron activation of the adjacent structures, and volumes are for a highly compacted hardware configuration. Even so, the densities are only 20 to 30 lb/ft³. This encourages the use of dense mass external

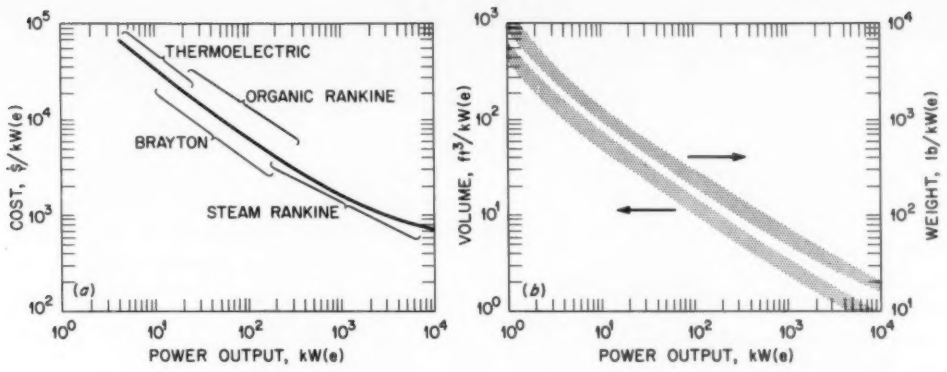

Fig. 2 (*a*) **Cost and** (*b*) **size of nuclear-reactor power plants as a function of power output.**

to the pressure hull for the dual functions of ballast and additional operational radiation shielding.

An umbilical power supply—a long extension cord—has been used successfully in a number of both near-shore and deep-sea missions. This concept has unique advantages and problems and is mentioned to indicate factors that might suggest further consideration. Costs for nominal surface and bottom conditions are strongly dependent on power level. Overall evaluation generally requires careful joint consideration of other mission factors—e.g., life support and communications.

MISSION PARAMETERS

Since the power system supports specific mission goals, evaluation must be according to parameters pertinent to the mission at hand. The cost of pressure hulls for containing the power system depends on the depth at which the system is to operate.

Charges must also be apportioned to the cost of delivery or resupply of the power plant and its fuel (if any)—the "elevator" charge. For items with a negative or neutral packaged net buoyancy, this is about $5 per pound—a round-trip charge since waste products must be removed from continuing resupplied missions—and is not greatly dependent on depth. The cost of delivering or resupplying at a surface point—the logistics rate—depends strongly on mission conditions and constraints—e.g., distance from supply port (or air base), prevailing local currents, sea state, weather, special handling procedures, and facilities required, criticality of delivery schedule, covert operations problems, and enemy interference. A rate of $500 per pound was recently specified as average for a U. S. Navy worldwide surface activity, and rates as high as several thousand dollars per pound have been experienced in difficult circumstances. For the present and near future, optimistic assumption of a logistics rate of no less than $10 per pound is suggested.

An assessment of manpower costs may be necessary where the power-plant operation requires human attention. Estimates range from $20 to $130 per deep-submergence man-hour, with $75 per hour reasonable for estimation purposes. Surface manpower will generally be accounted separately, where appropriate, and is much cheaper.

Temperature and heat-balance requirements are important selection factors in cases when process heat is available and needed. If cooling is required, costs of insulation and refrigeration due to the power-plant process must be considered.

Mission duration and total kilowatt-hour requirements must be considered. In addition, the maximum, minimum, and average power requirements and their time variations must also be taken into account.

CONCLUSIONS

The above considerations indicate that batteries are likely to be preferred as the prime power source for very short missions, where their relatively low cost overcomes other considerations. Fuel cells gain advantage over batteries with increasing mission duration because of their higher energy density. Nuclear power systems are preferred for intermediate- to long-duration missions, where extremely high energy density is of utmost importance. Reactor systems are preferred over isotopes, except at low power levels, due to the lower fuel and handling costs. (MG)

SNAP-21*

By F. Fox, R. Pannemann, and R. Wickenberg
(Summarized by Martha Gerrard)

Supplementary Keywords: *energy source; ocean; thermoelectric system;* ^{90}Sr.

The SNAP-21 (*S*ystems for *N*uclear *A*uxiliary *P*ower) system (Fig. 1) is a radioisotope-fueled thermoelectric generator designed for deep-sea applications. It converts the decay heat from a radioisotope into electric energy, using semiconductor thermoelectric materials maintained in a temperature gradient by means of seawater acting as the heat sink. This is a summary of USAEC Report MMM-3691-62.

SYSTEM DESCRIPTION

The radioisotopic heat source, $^{90}SrTiO_3$, is enclosed in a capsule designed to contain the fuel under all credible situations. It is surrounded by a two-piece shield made of U–8 wt.% Mo alloy, which decreases the radiation to a safe level. The shield, in turn, is surrounded by a thermal-insulation system except in the area where the flat-plate thermoelectric generator receives its heat. The thermal-insulation system consists of heat-reflecting foils and quartz-fiber spacers, wrapped around the biological shield and maintained under a high vacuum. A housing surrounds the entire assembly.

The generator for direct conversion of thermal energy to electric energy consists of two different types of semiconductor thermoelectric materials arranged in an electrical circuit and maintained in a temperature gradient. An electric potential is created in the circuit by the Seebeck effect, and, when the circuit is connected to a resistive load, a current will flow.

The thermoelectric generator is attached to a mounting plate, which also supports and locates the thermal-insulation system in the pressure-vessel body. An electric-power conditioner mounted in the pressure-vessel cover converts the electrical output of the thermoelectric generator to the desired voltage. On the cover of a two-piece pressure vessel enclosing all the components are mounted a receptacle and plug for electrical leads. A reusable handling, storage, and shipping container provides thermal and dynamic protection during transportation and storage.

Primary operating parameters of the system are

Power output	10 W(e)
Design life	5 years
Output voltage, d-c	24 ± 1 V
Load resistance	57.6 Ω
Maximum output voltage ripple, peak to peak	100 mV
Current at rated load	0.417 A

Performance is based on operation in 28 to 41°F seawater.

SAFETY SYSTEM

The safety of SNAP-21 is based on complete containment of the radioactive fuel under a maximum credible accident. The intrinsic safety of the system has been established through analysis of postulated situations that might jeopardize the integrity of the fuel capsule and by tests conducted on development fuel capsules. The results indicate that, under a maximum credible accident involving an aircraft crash accompanied by fire, the fuel would be contained. Evaluation of the consequences of fuel release if the maximum-credible-accident condition should be exceeded and of the effect of atmospheric and ocean fuel dispersion is presented in Report MMM-3691-49, Final System Safety Analysis. An operational safety analysis was also performed for long-term ocean implantment tests at San Clemente Island (Report MMM-3691-48).

Additional evaluation of specific mission hazards will be made for future operational systems to ensure that risk is minimized by identification of unique hazards and incorporation of appropriate safeguards.

OPERATIONAL PERFORMANCE

Three fueled systems have operated for about 2 years in the ocean at San Clemente Island off the coast of California. A fourth unit has been operating for a similar period under laboratory conditions. The units have operated stably and without degradation (other than fuel decay). Power output has exceeded design specifications.

*Original report title: SNAP-21 Program, Phase II. Deep Sea Radioisotope-Fueled Thermoelectric Generator Power Supply System, Final Summary Report 10-Watt System, May 1970, by Space and Defense Products, 3M Company, St. Paul, Minn. 55101.

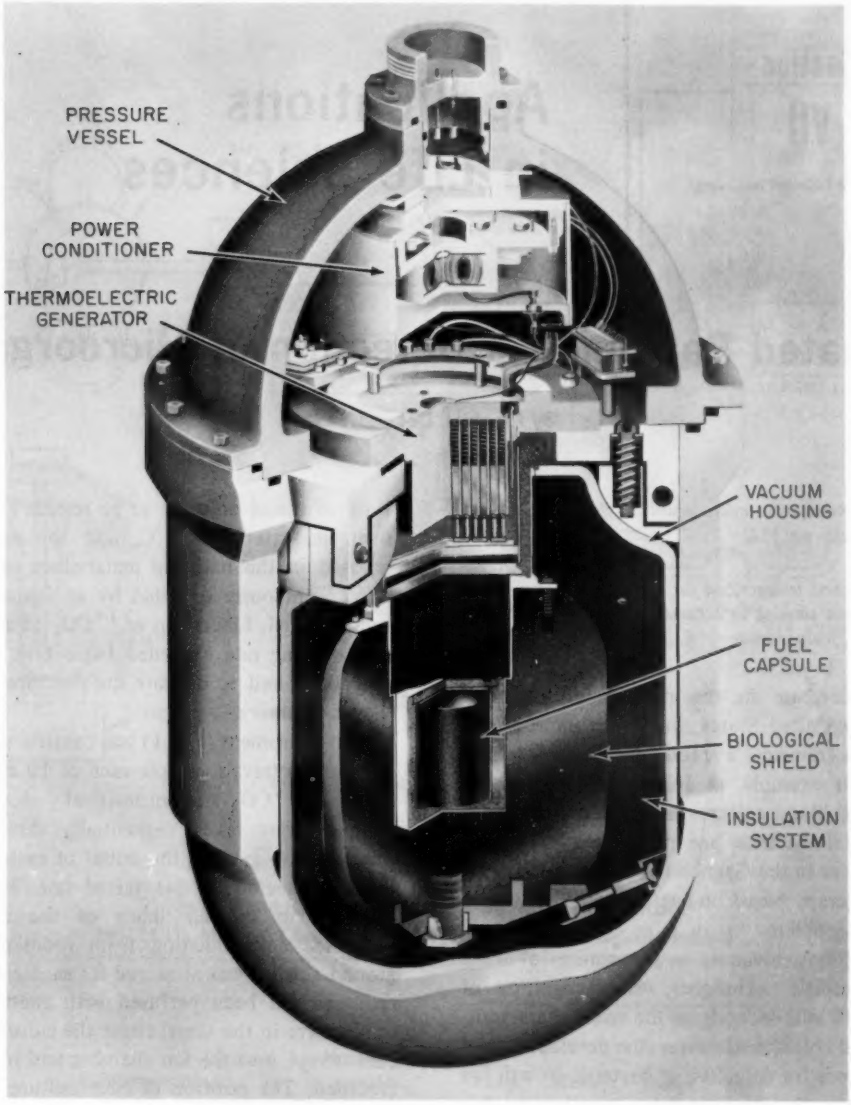

Fig. 1 SNAP-21 10-W system.

Section VII

Isotopes and Radiation Technology

Applications in Life Sciences

Automated Radiometric Detection of Microorganisms

By Frank H. DeLand, M.D.*

Supplementary Keywords: *microorganism; chemistry, analytical; medicine; diagnosis;* ^{14}C.

Abstract: *A method is described for detecting the presence of specific organisms causing infectious diseases, based on the release of* $^{14}CO_2$ *from glucose–*^{14}C.

Despite the decrease in the number of infectious diseases in the United States, the detection of microbial infections remains a problem of considerable magnitude. For example, at Johns Hopkins Hospital alone, 40 to 60 blood cultures each day are examined to determine if bacteria are present in the blood stream. Advances in the diagnosis of infectious diseases and specific therapy based on bacterial sensitivity have aggravated the problem.

Despite striking advances in automation of some types of diagnostic techniques, the recognition of bacterial growth still depends on the visual characteristics of bacterial colonies. However, the development of objective methods for detection of bacterial growth has been receiving appreciable attention recently, and this article describes a new method for the detection of bacteria by radiometric techniques that are readily automated.[1] The method, based on metabolism of glucose as an index of bacterial growth, is an extension of the work of Levin et al.,[2] who used radioisotope-labeled metabolites in an instrument for detecting life on Mars.

METHOD

The method consisted in inoculating a glucose-free fluid thioglycolate medium to which 0.5 μCi (36 μg) of uniformly labeled D-glucose–^{14}C had been added with 1 ml of blood or urine to be tested. The culture was then incubated at 37°C, and the rate of $^{14}CO_2$ released in the bacterial metabolism of glucose–^{14}C was continuously detected by an ionization chamber and recorded. Liberation of $^{14}CO_2$ at a rate such that the counting rate exceeded twice that of background was considered to indicate the presence of bacteria in the inoculum.

An instrument (Fig. 1) was constructed that would, at hourly intervals, sample each of 10 cultures for the release of $^{14}CO_2$ and automatically record the results. Samples were taken sequentially through a rotary-selector-valve system, the outlet of each culture vessel being connected to one valved line. Millipore filters attached to the air inlets of the culture vessels prevented contamination from room air. The background activity was measured for each culture after the chamber had been perfused with room air, and the atmosphere in the vessel above the culture was periodically swept into the ion chamber and its radioactivity recorded. The position of each culture in the instrument was recorded during each cycle.

RESULTS

Organisms, in concentrations of 10 to 100 individuals per culture, which have been detected by this method to date are *Aerobacter aerogenes,* anaerobic diphtheroids, *Diplococcus pneumoniae, Escherichia coli, Hemophilus influenzae, Herellea vaginicola, Proteus vulgaris, Salmonella typhosa, Shigella flexneri,*

*Division of Nuclear Medicine, The Johns Hopkins Medical Institutions, Baltimore, Md. Research supported in part by the U. S. Public Health Service.

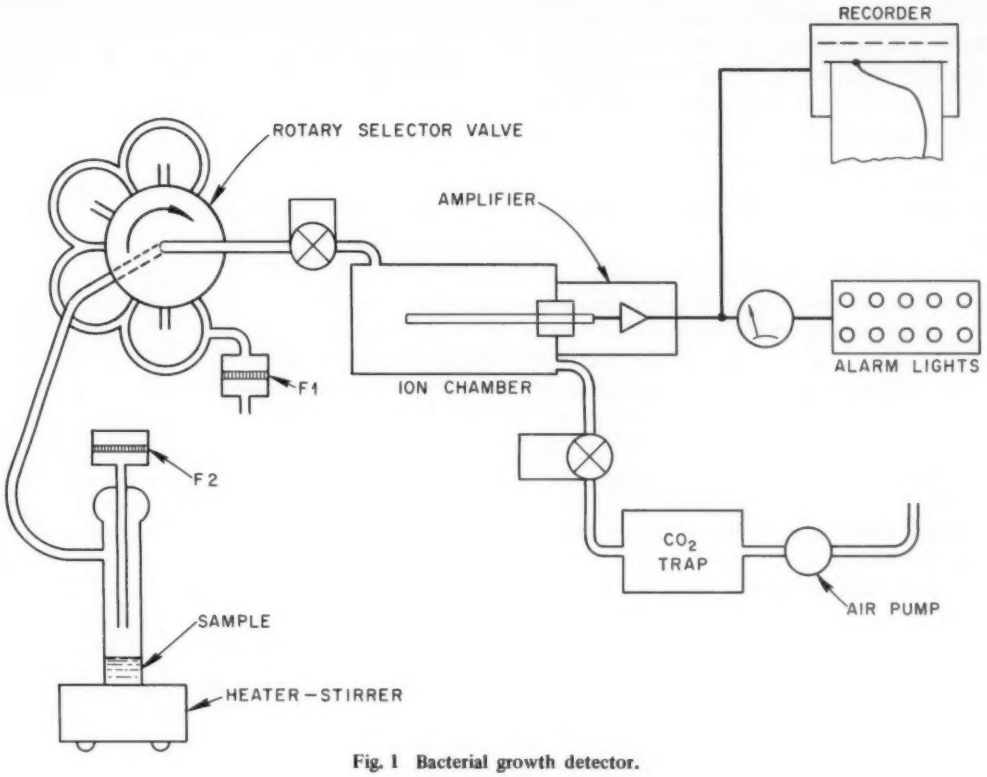

Fig. 1 Bacterial growth detector.

Table 1 Time Required for Detection of Microorganisms by $^{14}CO_2$ Method Compared with Routine Bacteriological Method

Microorganism	Number of samples	Time, hr			
		$^{14}Co_2$		Routine bacteriological	
		Range	Mean	Range	Mean
Diplococcus pneumoniae	8	1–5	1.7	24–48	36
Staphylococcus aureus	6*	1–6	3.3	24	24
Micrococcus species†	1	1	1	24	24
Streptococcus fecalis	1	1	1	24	24
Proteus vulgaris	3*	1–2	1.3	24	24
Escherichia coli	4	1–4	3.0	24	24
Aerobacter aerogenes	3	1–2	1.3	24	24
Bacillus subtilis†	2	1	1	24	24
Candida albicans	2	21–27	24	48	48

*By routine bacteriological methods *Staphylococcus aureus* was not recovered from three blood samples of one patient nor was *Proteus vulgaris* from one sample from another patient.

†Probably not of clinical significance.

Staphylococcus aureus and *epidermidis*, beta and alpha hemolytic streptococci, and *Streptococcus fecalis*.

The method was developed with pure cultures from the Bacteriology Laboratory of the Johns Hopkins Hospital. The time required for positive identification of growth was generally 1 to 6 hr—from a half to a twentieth of that required by routine methods (Table 1, preceding page). The amount of ^{14}C recovered as $^{14}CO_2$ varied from 16 to 18% of that added as glucose–^{14}C.

The solubility of CO_2 in an aqueous solution at *p*H 7 is quite high, and the gas remains in solution until the *p*H of the culture medium has decreased to about 5. To identify the production of $^{14}CO_2$ as early as possible, the medium was kept in constant agitation with magnetic stirrers. This facilitated physical exchange between $^{14}CO_2$ in solution with the stable CO_2 in the atmosphere above the medium, which occurs even when the *p*H of the culture medium is 7. Without agitation, it took three to four times as long to detect bacterial metabolism.

In more than 300 positive cultures of bacteria of 18 different species, we have found that $^{14}CO_2$ is released by the bacterial metabolism of D-glucose–^{14}C in amounts adequate for detection by radiation instrumentation. There have been no false positive results from 600 negative cultures even though each contained at least 1 ml of whole blood. Release rates are similar for most organisms (Fig. 2), especially during the first hour. This suggests that during the early stage of incubation the number of viable bacteria remains constant. The number of bacteria present at 2 hr, as determined by colony counts on blood-agar culture plates, is essentially the same as in the inoculum, and the more rapid release of $^{14}CO_2$ coincides with multiplication of the bacteria. If the bacteria utilize the glucose only in a pathway that yields one molecule of CO_2, then the recovery represents 100% of the glucose. However, more than one metabolic pathway is often involved, e.g., a system in which each molecule of glucose forms two pyruvate units, each of which may yield CO_2. On this basis, recovery should be 50% or less for most bacteria. We found a rate of 16 to 18% during this lag phase of bacterial multiplication, during which both the average cell size and the metabolic activity of each cell increase. During the first 2 hr of incubation, an ion chamber is quite adequate for detecting the metabolism of less than 100 bacteria. The sensitivity of the system might be increased severalfold by decreasing the volume of the 1.5-liter system.

The automated bacterial growth detector is now being evaluated in the hospital bacteriological laboratory. To date, 5 hundred blood specimens from patients in the Johns Hopkins Hospital have been studied with this instrument, and the results compared with routine bacteriological methods. As detected by $^{14}CO_2$ release, 30 of these specimens were positive for bacterial growth. There were no false negative results, although four of the cultures shown to be positive by the radiometric technique were negative by routine bacteriological methods. (MG)

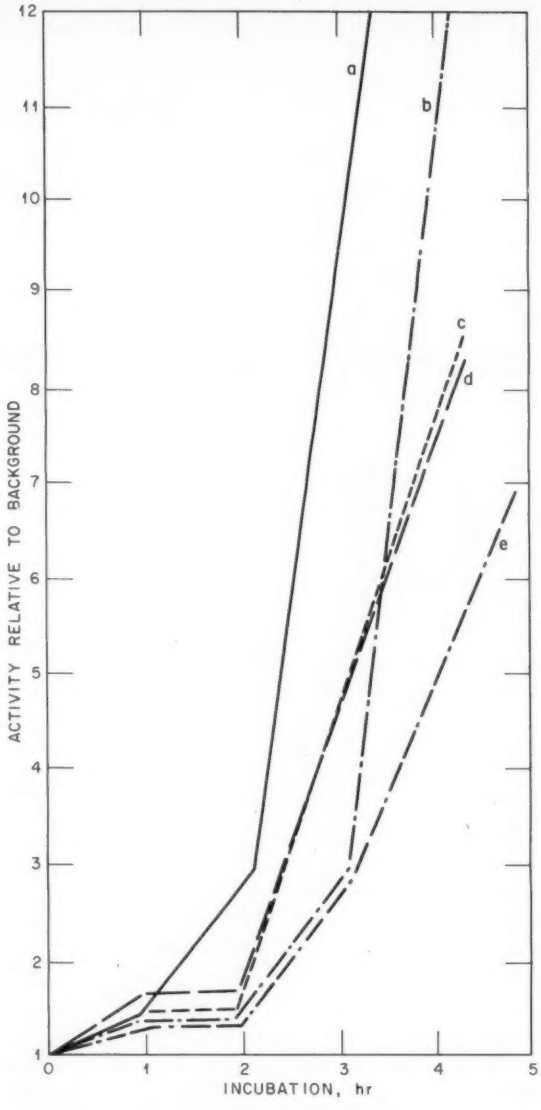

Fig. 2 Release of $^{14}CO_2$ from labeled glucose by (*a*) *Streptococcus fecalis*, (*b*) *Aerobacter aerogenes*, (*c*) *Salmonella typhosa*, (*d*) *Escherichia coli*, and (*e*) *Staphylococcus aureus*.

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A TRAINING MANUAL FOR NUCLEAR MEDICINE TECHNOLOGISTS

By Guy H. Simmons, Division of Medical Radiation Exposure, Bureau of Radiological Health, Cincinnati, Ohio, October 1970, Report BRH/DMRE 70-3 of U. S. Department of Health, Education, and Welfare, Public Health Service, Bureau of Radiological Health, Rockville, Md. 20852.

This manual was prepared for the training program in Nuclear Medicine Technology at the University of Cincinnati. Introduced by a basic discussion of atomic structure and radioactivity, its contents include a discussion of laboratory counting systems, principles of detection and of in vivo and in vitro counting, radioactive tracers and pharmaceuticals, and clinical diagnostic studies, together with descriptions of scanning and radiation monitoring instruments. Appendixes include background material. (MG)

STERILE-MALE TECHNIQUE FOR CONTROL OF FRUIT FLIES

(Proceedings of a Panel, Sept. 1–5, 1969, organized by the Joint FAO/IAEA Division of Atomic Energy in Food and Agriculture), IAEA, Vienna, 1970 (STI/PUB/276) (175 pp., 28 papers; $5.00).

Supplementary Keywords: *source, gamma; ecology; entomology; insect control; material tracing; bibliography;* ^{32}P; ^{60}Co; ^{137}Cs.

The sterile-male technique of insect control and eradication is especially effective on the fruit-fly family *Tephritidae*, because they can be raised cheaply in large numbers. Thus the technique—chiefly irradiation with gamma rays from ^{60}Co or ^{137}Cs—has been used more extensively with fruit flies than with any other insect pests. Developing countries plan to extend the evaluation to other fruit-fly species. Projects in Central America (site of the largest international investment in research and development), Italy, and Spain show that much biological and ecological data are needed before the method can be applied on a large scale. Some other fruit-fly species (e.g., Mediterranean and Mexican fruit flies and melon fly) can also be raised inexpensively, but complete mechanization must be achieved before large-scale eradication is begun. For several other species, none of the basic data needed are available.

This publication summarizes a panel meeting of 24 participants from 12 countries; it contains 12 short communications on work done at various institutions plus 16 formal reports. Papers on the Mediterranean fruit fly outnumber those on other species and are grouped separately. Information is given about mass-rearing techniques, fruit-fly physiology and ecology, comparison of techniques for control and eradication, and the status of programs in progress. The general conclusion is that the sterile-male technique will find increasing application, especially since it holds no dangers for man.

The recommendations are a very important part of the book. The panel points out the need for more information on the basic biology, physiology, and ecology of the fruit fly. It suggests the adoption, by national and regional organizations, of the methods and approaches developed by international organizations; further, it recommends the establishment of one or more International Fruit Fly Research and Training Centers. It gives specific recommendations on research programs for sterile-male-technique control and eradication of five fruit-fly species. Included is a detailed outline of a proposed large-scale experiment with this technique to eradicate from Nicaragua the Mediterranean fruit fly. The tremendous economic losses that could result if the fruit fly spreads still further from Nicaragua—as it might well do—more than justify implementation of the experiment.

The publication is an excellent source book for those engaged in programs to control or eradicate fruit flies or other insects. (HPR)

Radioisotopic Technique for Studying Biological Slime-Treatment Processes

By George W. Reid*

Supplementary Keywords: *pollution; water and waste treatment; apparatus; tracer;* ^{32}P.

Abstract: *Apparatus for studying the effect of variables in slime treatment of sewage was constructed. Slime is deposited on a drum that rotates in a medium labeled with* ^{32}P, *and the activity of the drum is counted to determine the uptake of phosphorus by the slime.*

Before sewage is discharged to natural waterways, it is screened to remove gross solids, allowed to settle and/or filtered to remove fine solids, and chlorinated to destroy bacteria. If the organic content is large, secondary treatment, for example with activated sludge, or even tertiary treatment to remove nitrogen- and phosphorus-containing nutrients is necessary to prevent depletion of dissolved oxygen and eutrophication in the receiving stream. Any of the compounds or radicals formed from the organics—CO_2, H_2O, NH_3, SO_4^{2-}, or PO_4^{3-}—may be used to follow the process and thus determine its effectiveness. If material containing ^{32}P is added to the substrate, the uptake of this material by the slime can be readily detected with a simple counting device.

The apparatus described here, in which a slime is alternately exposed to air and to a labeled medium, was based on studies made, under a USAEC contract, on the possible radiation danger of accumulated ^{32}P in domestic plumbing and on levels of radioactivity in natural waters.[1]

INSTRUMENT

The apparatus (Fig. 1) consists of a solid glass or Plexiglas cylinder, 10 cm in diameter by 5 cm long, whose surface has been frosted by sandblasting. It is mounted on a central shaft, which is rotated by a geared-down electric motor (the latest design uses a rotisserie spit and motor) and is partly submerged in a medium containing ^{32}P. Growth of the slime is started by initially seeding the substrate—nutrient broth or beef extract and sugar—with raw sewage. The pans containing the substrate are changed frequently until a thick slime is obtained, which usually requires about 48 hr. As the cylinder rotates, the microorganisms forming the slime take up the ^{32}P, the slime deposited on its surface is alternately exposed to oxygen and substrate, and the accumulation of ^{32}P by the slime is measured by a detector located over the cylinder. The rate of uptake increases rapidly for the first 10 to 15 min, being due chiefly to absorption of the medium during this period. Subsequent uptake is a combination of absorption and assimilation. The radioactivity may be recorded continuously or at hourly intervals.

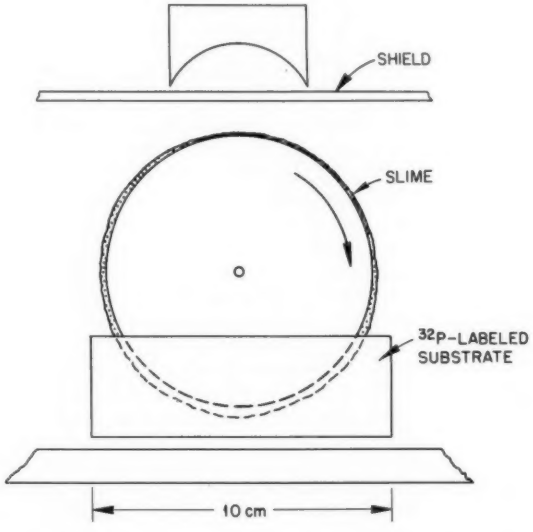

Fig. 1 Rotating-drum apparatus for studying slime metabolism.

The substrate container should be made of a nontoxic material, e.g., glass or aluminum, and the best size was a 2- by 14- by 23-cm pan. When the organic concentration is very high, the pan should be fitted with a larger reservoir, e.g., a 20-liter carboy, and a circulating pump. The detector, a 4.5 mg/cm^2 glass G-M tube, was not affected by slime thickness, but the

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detector aperture was carefully controlled. A cylinder rotation speed of 7 to 10 rpm and an initial ^{32}P concentration of 3 μCi/liter have been found satisfactory. Higher concentrations tend to overload the detector and smaller ones to introduce statistical problems.

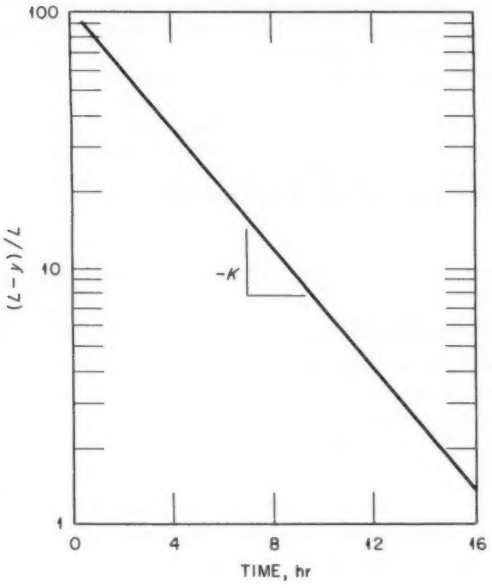

Fig. 2 Rate of ^{32}P uptake by slime.

The rate at which the treatment is progressing is determined from

$$\frac{dy}{dt} = -k(L - y), \quad \text{from which} \quad \log\frac{L - y}{L} = -kt,$$

where y is the phosphorus uptake at time t, L the ultimate uptake or impressed load, and k the slope of the straight line obtained when the logarithm of the term $(L - y)/L$ is plotted vs. t.

In 12 to 14 hr, equilibrium of the tracer is established, after which the accumulated activity may decrease. The value of k is then determined from the plot of $(L - y)/L$ vs. t (Fig. 2), using the highest uptake value found as L and all others as y. The substrate can have additional ^{32}P added, and another series can be overlaid on the same slime. Values of k are reproducible ($R^2 \geqslant 0.98$) where similar substrate concentrations are used, but a decrease in pH is normal throughout an experiment. (MG)

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TRACER PROBES IN STEADY-STATE SYSTEMS

By Robert Steele (Brookhaven National Laboratory), Charles C Thomas, Publisher, Springfield, Ill., 1971 (236 pp.; $17.00).

Although radioisotopic tracers have been widely used to determine chemical pathways in metabolism, the technique has been less readily adapted to the measurement of metabolic rates. Rate data are not too difficult to obtain, but interpretation is not straightforward because of meager knowledge of the systems and thus the unsuitability of the mathematical models applied.

Steele's book presents the basic principles on which tracer measurements of metabolic rates are based. The mathematical development is designed to be understood by persons specializing in fields other than mathematics, with practical use of the material as the final objective. The procedures are illustrated with specific numerical systems from mammalian metabolism experiments. Well-mixed metabolic pools, precursor–product relations, one-compartment models, two-compartment models, complex systems, and continuous infusion of tracer material are among the concepts considered. The principles discussed should be applicable to fields other than metabolic rate determinations, e.g., in tracing elements in a near-steady-state ecosystem. (MG)

DYNAMIC STUDIES WITH RADIOISOTOPES IN MEDICINE

Symposium Proceedings, Rotterdam, Aug. 31–Sept. 4, 1970, IAEA, Vienna, 1971 (STI/PUB/263).

Progress in applications of radioisotopes to clinical diagnosis was discussed at the Rotterdam Conference by 320 participants from 37 countries. Subjects covered included newly available radioisotopes, new instruments, new techniques, and new methods of data analysis. The 70 papers presented include several extensive and interesting reviews—all related to radioisotope usage, of course: theoretical aspects of dynamic clinical studies and studies in thyroid, renal, lung, and liver functions; iron metabolism; regional blood flow; and quantitative radiocardiography. Both procedures that have come into routine usage and procedures still under development are discussed. Of special interest in the latter group are papers on quantitative analysis of scintillation camera data, the great interest in which is attested to not only by the number of papers presented but also by the discussion that followed. This book is valuable as an indication of the present status of this field. (MG)

ECONOMICS OF SEED IRRADIATION

The USSR's 1300-Ci ^{137}Cs Kolos ("Ear of Corn") unit for presowing irradiation of seed was described at the Nuclex 69 exhibit.* A mobile production model unit, 3470 Ci of ^{137}Cs, has been constructed which can irradiate 1000 kg of seed per hour with a dose of 800 to 1000 rads and a radiation utilization efficiency of 22%. Preliminary test data† indicate that crop yields are increased from 10% for winter wheat to 25% for sugar beets and carrots. Tentative estimates indicate that irradiation costs will be recovered in one season. Quantity production of the unit is planned. (MG)

*Sealed Radioisotopic Sources at Nuclex 69, *Isotop. Radiat. Technol.*, 7(4): 459-460 (Summer 1970).

†N. S. Prokof'ev, D. A. Kaushanskii, and B. G. Zhukov, *At. Energ. (Moscow)*, **28:** 453-454 (1970).

IAEA SCHEDULES SYMPOSIUM ON NUCLEAR ACTIVATION TECHNIQUES IN THE LIFE SCIENCES

Ljubljana, Yugoslavia, has been selected as the site for an IAEA-sponsored symposium on Nuclear Activation Techniques in the Life Sciences, Apr. 10–14, 1972. This a a follow-up to the May 1967 Amsterdam symposium on the same subject.

Four general subject areas have been selected:

I. *Physical and Chemical Procedures,* which includes sample-preparation techniques, recent advances in activation techniques, automated as well as multicomponent and group separation sciences, comparisons of activation with other trace-element assay methods, and preparation and use of biological analytical reference material.

II. *Biology,* which includes cellular and subcellular system studies and microbiology, plant and agricultural studies (e.g., ecology, deficiency states, accumulation phenomena), and animal studies (e.g., ecology, deficiency states, accumulation phenomena).

III. *Medical and Dental Sciences,* including biochemical and physiological studies (e.g., of essential trace elements, by the use of enriched stable isotopes), applications in clinical and diagnostic research and in therapy (e.g., studies of trace elements in blood and in body tissues), in vivo activation-analysis techniques, and pharmacological and pharmaceutical studies (e.g., drug-quality determination and control).

IV. *Human Ecology,* comprising public health and environmental pollution (e.g., studies of trace elements in air and in water), and food additives and contamination.

Proceedings will be published by the IAEA. Questions concerning the meeting should be directed to Mr. H. H. Storhaug, Division of Scientific and Technical Information, International Atomic Energy Agency, Kärntner Ring 11, P. O. Box 590, A-1011 Vienna, Austria.

Section
VIII
Isotopes and Radiation Technology

Miscellaneous

Availability of Isotopes and Services

NEUTRON-SOURCE SHIPPING CONTAINER

The availability of shielded shipping containers that meet U. S. Department of Transportation shipping regulations for neutron sources is announced by NuTex Corporation (P. O. Box 13124, Capitol Station, Austin, Tex. 78711). This steel-encased Cal-Shield is made of a boron-impregnated water-extended polyester resin and is less expensive than containers made from solid hydrocarbons, such as paraffin. These containers can be used for ^{252}Cf as well as for Po–Be, Pu–Be, or Am–Be neutron sources. The basic price for a container to be used with a 1-mg ^{252}Cf source (2.3×10^9 n/sec) is $725.

CALIFORNIUM-252 NOW AVAILABLE

An encapsulated ^{252}Cf source was on display at the United Nations Conference on Peaceful Uses of Atomic Energy in Geneva in September 1971. These sources are now available from five U. S. companies: General Electric's radiation processing group, in Pleasanton, Calif.; Gamma Industries' Division of Nuclear Systems, Inc., Baton Rouge, La.; Sanders Nuclear Corp., Nashua, N. H.; Nuclear Materials and Equipment Corp., Apollo, Pa.; and Monsanto Research Corp.'s Nuclear Products Department, Dayton, Ohio.

The first purchaser of encapsulated ^{252}Cf after the USAEC offered this nuclide for sale was the University of Kentucky, who paid the AEC $13,500 for the 1-mg source. Sale of bulk (unencapsulated) ^{252}Cf by the AEC started in February 1971.

Reactor Experiments, Inc. (963 Terminal Way, San Carlos, Calif. 94070), has announced the fabrication of casks shielded with a borated hydrocarbon material that will package a 10^9-n/sec ^{252}Cf source.

COBALT-60 LOAN PROGRAM

The USAEC is conducting a ^{60}Co loan program to foster research and development on the use of ^{60}Co as a heat source (see article on ^{60}Co heat sources in this issue). Cobalt-60 with specific activities of 200 to 500 Ci/g in the form of wafers, half-wafers, and slabs will be loaned without charge, f.o.b. Savannah River Laboratory, to participants, who are expected to fund their own work. The results of the research will be made available to the AEC, but patent arrangements will be made to reflect the relative contributions of the borrower and the AEC.

The loaned ^{60}Co may not be used in such a way as to compromise marketing of lower specific activity ^{60}Co by commercial producers.

Further information concerning the conditions of the program and submission of proposals can be obtained from

Manager, Savannah River Operations Office
U. S. Atomic Energy Commission
P. O. Box A
Aiken, S. C. 29801
Attention: N. J. Donahue
803-824-6331, Ext. 2623

AEC Activities

CONTRACTS FOR ARTIFICIAL HEART SYSTEMS

The AEC is negotiating contracts with TRW Systems of Redondo Beach, Calif., and with Westinghouse Astronuclear Laboratory, to evaluate the practicability of a radioisotopic thermal converter for an artificial heart device. TRW will continue the project with development of a ^{238}Pu heat source.

EXEMPTION FOR STATIC-ELIMINATION DEVICES AND ION-GENERATING TUBES

The AEC is proposing to exempt from licensing the use of static-elimination devices and ion-generating tubes containing ^{3}H, ^{85}Kr, or ^{210}Po, items now used under general licenses. The proposed amendments also establish requirements for specific licenses to manufacture, distribute, import, or incorporate such devices into products for commercial distribution. Under the new regulations, a manufacturer seeking a specific license for such products would be required to submit information about the design, manufacture, prototype testing, quality-control procedures, and labeling which would demonstrate that the product would not release the radioactive material or its radiation under either normal or severe conditions of handling, storage, use, and disposal.

Further information may be obtained from the Secretary of the Commission, U. S. Atomic Energy Commission, Washington, D. C. 20545, Attention: Chief, Public Proceedings Branch.

General

ISOTOPIC CARDIAC PACEMAKERS APPROVED IN EUROPE

Seven European countries—France, West Germany, Spain, Sweden, Switzerland, Austria, and Denmark—have signed an agreement allowing free travel of persons wearing radioisotopic cardiac pacemakers. At an ENEA (European Nuclear Energy Agency) meeting in Paris in May, which included delegates from Britain, Canada, and the United States, broadening of the agreement and legal and medical aspects of pacemaker-patient travel were discussed.

The fourth Frenchman to receive a nuclear pacemaker implant is doing well, and 25 to 50 more such implants are expected before the end of 1971.

NEW AUSTRALIAN IRRADIATION PLANT

Australia's first gamma-irradiation plant designed specially for medical purposes is being built at Dandenong, Victoria, by Tasman Vaccine Laboratory (Australia) Pty. Ltd. The $1 million undertaking will sterilize medical packs for hospitals throughout Australia. Equipment is being installed by Atomic Energy of Canada Limited.

The complex is designed to meet the most stringent safety standards. It has walls 6 ft thick and a roof 4.5 ft thick. More than 1000 tons of special concrete were poured continuously to ensure a perfect, fault-free bond. Start-up was scheduled for July 1971 with the fully automated and electronically controlled plant operating 24 hr/day, 7 days/week.

NEUTRON-SOURCE CONFERENCE

The American Nuclear Society National Topical Meeting, *Neutron Sources and Applications*, in Augusta, Ga., April 19–21, 1971, attracted some 250 persons from the United States and abroad. The program consisted of 19 invited papers, largely the rapporteurs' summaries of the contributed papers, which had been sent to them in advance, and one panel discussion. These, together with comments from the floor and the approximately 70 contributed papers on reactor, accelerator, and isotopic neutron sources, are printed in Vols. I to III of the proceedings (Report CONF-710402).

GENETIC IMPROVEMENT OF INDUSTRIAL MICROORGANISMS

Genetic alterations in and selections of microorganisms that are or may be valuable in the large-scale production of medicinal compounds such as antibiotics and of foodstuffs were reviewed at an IAEA symposium in Vienna in March. The approximately 100 participants—from 27 countries and six international or regional organizations—included microbiologists, biochemists, and geneticists.

SCREWWORM BARRIER

In "Facts About the Screwworm Barrier Program" (Report ARS 91-64-1, October 1969), the U. S. Department of Agriculture (Animal Health Division, Agricultural Research Service, Federal Center Building, Hyattsville, Md. 20782) describes the screwworm content in a 300- to 500-mile-wide zone along the 2000-mile-long U. S.–Mexico border. The pest has been eradicated in the United States, and periodic release of sterile males in or near this barrier zone prevents reestablishment of the problem. The sterile flies are produced at a rate of 135 million (50% males) per week by ^{60}Co irradiation of 5.5-day-old pupae.

IAEA TO SPONSOR TWO SYMPOSIUMS

The IAEA will sponsor a symposium, "Dosimetry Techniques Applied to Agriculture, Industry, Biology, and Medicine," in Prague, Czechoslovakia, Apr. 17–21, 1972. Papers are expected to be presented on instrumentation and techniques and effects of neutron, gamma, X, and charged-particle radiation and will include reports on calorimetry; various types of dosimetry, such as chemical, film, radiophotoluminescent, and thermoluminescent; dosimetric systems; and liquid dielectric ionization chambers. The Scientific Secretaries will be Messrs. W. S. Moos and L. Chandler, Division of Life Sciences, International Atomic Energy Agency, Kärntner Ring 11, P. O. Box 590, A-1011, Vienna, Austria.

The Agency and the Food and Agriculture Organization of the United Nations will also cosponsor a symposium, "The Use of Isotopes in Studies on the Physiology of Domestic Animals with Special Reference to Hot Climates," in Athens, Greece, Mar. 20–24, 1972. Topics to be covered include problems of nutrition and digestion in hot climates, water and salt metabolism, tracers in body composition studies, hormonal control of metabolic rate and heat production, and endocrine aspects of reproduction. The Scientific Secretary is Mr. H. A. Höller, Joint FAO/IAEA Division of Atomic Energy in Food and Agriculture, located at the IAEA address given above.

Participation in both symposiums will be by persons nominated by the government of a member state of the sponsoring organization(s) or by an international organization invited to participate. The working languages of the meetings will be English, French, Russian, and Spanish, and all communications, abstracts, and papers must be submitted in one of these languages.

PLASTICS-IN-PACKAGING CONFERENCE

The Ninth Washington State University conference on "Plastics in Packaging" will be held on June 21–23, 1972. For information, contact the Engineering Extension Service, Washington State University, Pullman, Wash. 99163.

USAEC publications of special interest

INDEX TO CONFERENCES RELATING TO NUCLEAR SCIENCE

Editors: Willie E. Clark, Doris H. McGinnis, and Carol B. Patterson

A valuable aid in identifying and locating conferences on nuclear science and technology, this index provides access to information on conferences covered in *Nuclear Science Abstracts*, January 1966 through June 1970 (Vol. 20, No. 1, through Vol. 24, No. 12), and indicates the conferences for which the Division of Technical Information Extension, USAEC, can provide availability information not readily found elsewhere. Conferences are indexed by dates, places, conference numbers, report numbers, and significant words in the titles. Volume and abstract numbers are included for conferences abstracted in *Nuclear Science Abstracts*. This publication updates TID-4043 (Rev. 3), which covers *Nuclear Science Abstracts* Vols. 11 through 19 (1957–1965), and supersedes TID-4043 (Rev. 3, Supplement). 369 pages.

CONTENTS

Part I: Permuted Titles and Cities

Part II: Numbered Conferences

Part III: Report Numbers

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Available as TID-4044 for $6.00 from
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Springfield, Virginia 22151

ENGINEERING MATERIALS LIST

Editors: Richard J. Smith, Francis L. Sachs, and Willie E. Clark

This series announces and describes packages of engineering materials made available to the public by the U. S. Atomic Energy Commission. These materials—drawings, specifications, design criteria, flow sheets, parts lists, bills of materials, photographs, maps, etc.—deal with individual instruments, processes, facilities, and items of equipment developed or investigated by AEC contractors. As they are made available at the National Technical Information Service, the packages are abstracted and indexed in periodic supplements to *Engineering Materials List* (TID-4100). The supplements and annual cumulative indexes to the supplements are available without charge upon request to the Division of Technical Information Extension, P. O. Box 62, Oak Ridge, Tennessee 37830.

Announcing the publication of

Public Safety & Underground Nuclear Detonations

Samuel Glasstone

June 1971

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The United States program for testing nuclear explosive systems has two main objectives: first, to maintain the nation's security and, second, to develop peaceful applications of nuclear explosives for the benefit of all mankind.

To meet the national objective of developing nuclear explosives for peaceful applications, the Atomic Energy Commission in 1957 established the Plowshare Program. For such applications as stimulating the flow of natural gas and petroleum, mining and quarrying and other earth-moving projects, and cracking high-temperature rocks to utilize their natural geothermal energy, the Plowshare Program will require the design and testing of special nuclear explosives.

This book describes the comprehensive safety program carried out by the Atomic Energy Commission and the methods through which those responsible for ensuring safety in underground nuclear detonations implement their basic philosophy, which has been summarized as follows: A nuclear device can be detonated safely only when it is ascertained that the operation can be accomplished without injury to people and domestic animals, directly or indirectly, and without an unacceptable damage risk to ecological systems and to natural and man-made structures.

Published by the Atomic Energy Commission, the book was prepared through the coordinated effort of government agencies and contractors; the work of many competent scientists in a number of related disciplines is presented.

AEC Symposium Series

neutron standards and flux normalization

Proceedings of a symposium held at Argonne National Laboratory, Argonne, Illinois, October 21–23, 1970

Coordinator: Alan B. Smith, Argonne National Laboratory

August 1971
540 pages, 6 x 9
Library of Congress Catalog Card: 77-611328

This symposium, sponsored by the European–American Nuclear Data Committee, emphasizes the need for precise nuclear quantities of critical importance to nuclear-energy programs, primarily reactor development. The symposium (1) quantitatively assesses the status of the field, (2) defines available precisions, (3) identifies outstanding problems, (4) formulates recommendations, and (5) provides guidance for future work.

The areas covered include

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Published as No. 23 in the AEC Symposium Series,
this book is available as CONF-701002 for $6.00 from
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NUCLEAR SCIENCE ABSTRACTS

The U. S. Atomic Energy Commission, Division of Technical Information, publishes *Nuclear Science Abstracts (NSA)*, a semimonthly journal containing abstracts of the literature of nuclear science and engineering.

NSA covers (1) research reports of the U. S. Atomic Energy Commission and its contractors; (2) research reports of government agencies, universities, and industrial research organizations on a worldwide basis; and (3) translations, patents, books, and articles appearing in technical and scientific journals.

Complete indexes covering subject, author, source, and report number are included in each issue. These indexes are cumulated and sold separately.

Availability

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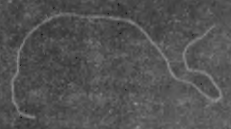
EXCHANGE *NSA* is also available on an exchange basis to universities, research institutions, industrial firms, and publishers of scientific information. Inquiries should be directed to the Division of Technical Information Extension, U. S. Atomic Energy Commission, P. O. Box 62, Oak Ridge, Tennessee 37830.



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